



## Mass spectrometry for Determination of Environmental Radionuclides

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# ENVIRA 2015

International Conference

Environmental  
Radioactivity

21-25 SEPT.

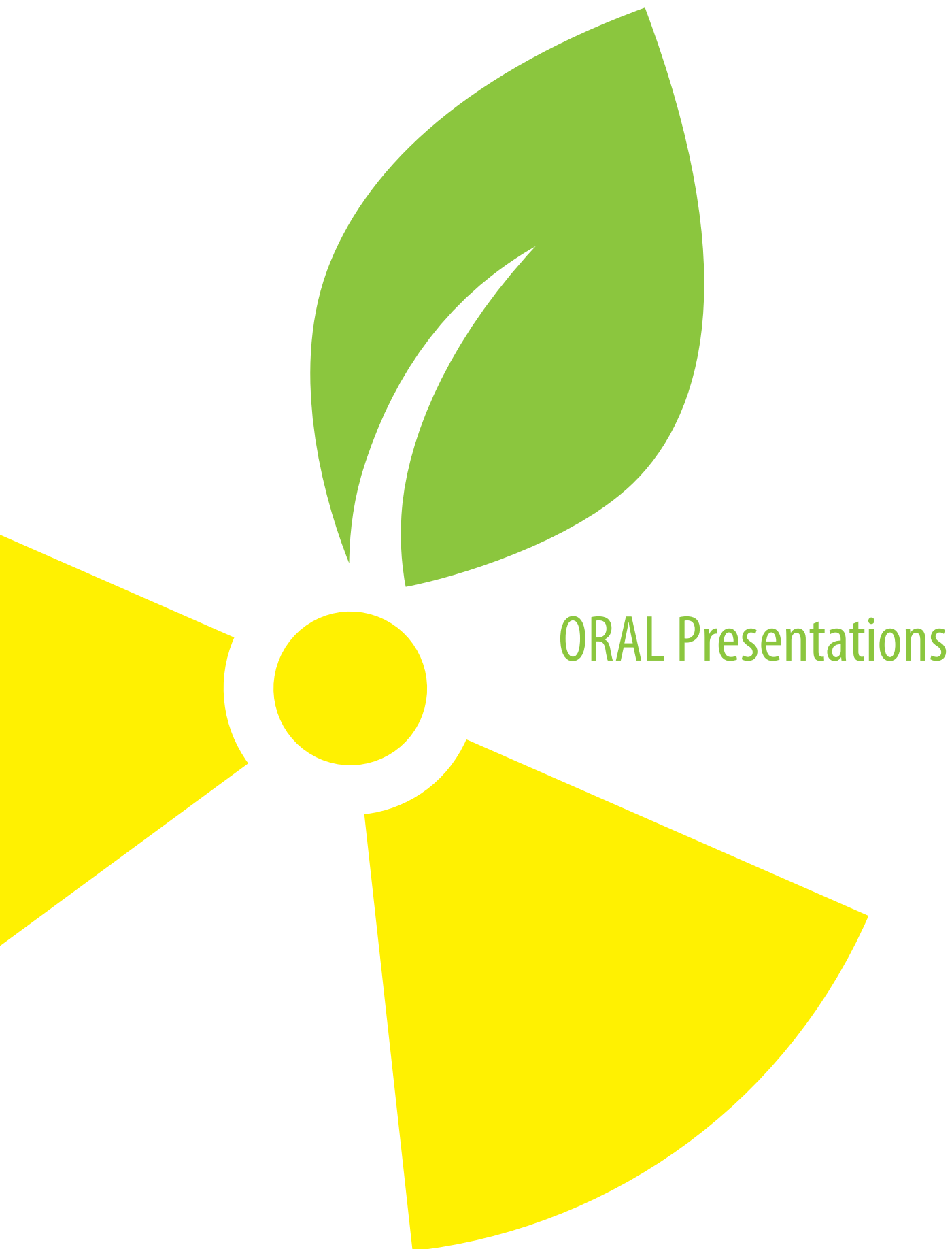
THESSALONIKI GREECE

**The MET hotel**

**CONFERENCE Proceedings**







ORAL Presentations

08:00-09:00 Registration

09:00-09:30	<b>ENVIRA CONFERENCE OPENING</b> <b>ROOM: MAISTROS A, CHAIR: A. IOANNIDOU</b>  Giannis Boutaris Mayor of Thessaloniki  Theodore Laopoulos Vice Rector for Research & Coordination, Professor of the School of Physics, Faculty of Sciences  George Kitis Vice President of the School of Physics, Faculty of Science  Christos Housiadas Chairman of GAEC, Address by the EEAE Chairman - Evolutions in the European regulatory framework for environmental radioactivity
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09:30-11:00 PL1	<b>PLENARY SESSION 1</b> <b>ROOM: MAISTROS A, CHAIR: P.P. POVINEC</b>
09:30-10:00 PL1-1	Peter Steier, Austria <b>NEW FRONTIERS FOR ACCELERATOR MASS SPECTROMETRY - NEW ISOTOPES AND NEW APPLICATIONS</b> <i>Peter Steier, Michaela Fröhlich, Johannes Lachner, Martin Martschini, Johanna Pitters, Francesca Quinto, Aya Sakaguchi, Stephan Winkler and Robin Golser</i>
10:00-10:30 PL1-2	Timothy Jull, USA <b>APPLICATIONS OF <sup>14</sup>C AND <sup>129</sup>I TO SOME PROBLEMS IN ENVIRONMENTAL SCIENCE</b> <i>Timothy Jull</i>
10:30-11:00 PL1-3	Walter Kutschera, Austria <b>ISOTOPES AND CLIMATE CHANGE</b> <i>Walter Kutschera</i>

11:00-11:30 Coffee Break

PARALLEL SESSIONS 1 11:30-13:00

<b>SESSION 1A: AMS</b> ROOM: MAISTROS A CHAIR: W. KUTSCHERA & W. KIESER	<b>SESSION 1B: MARINE</b> ROOM: SIROCCO CHAIR: S. CHARMASSON & H. FLOROU
11:30 O1 A-1  Haruka Kusuno, Japan <b>COMPARISON OF <sup>129</sup>I/<sup>127</sup>I RATIO IN FISH AND SEAWATER SAMPLES FROM THE PACIFIC OCEAN</b> <i>Haruka Kusuno, Hiroyuki Matsuzaki, Toshi Nagata, Yosuke Miyairi, Yusuke Yokoyama, Naohiko Ohkouchi</i>	11:30 O1 B-1  Sang-Han Lee, Korea <b>THE TRANSPORT OF CLOSE-IN FALLOUT PLUTONIUM IN THE NORTHWEST PACIFIC OCEAN : TRACING THE WATER MASS MOVEMENT USING <sup>240</sup>Pu/<sup>239</sup>Pu ATOM RATIO IN SEDIMENT</b> <i>Sang Han Lee, Jung Suk Oh, Gi Hoon Hong, Moon-Sik Suk, Chul-Soo Kim and Pavel Povinec</i>
11:45 O1 A-2  Angel T. Bautista Vii, Japan <b>IODINE-129 IN CORAL CORES FROM THE PHILIPPINES AS A TRACER OF ANTHROPOGENIC NUCLEAR ACTIVITIES</b> <i>Angel T. Bautista Vii, Hiroyuki Matsuzaki, Fernando P. Siringan</i>	12:00 O1 B-2  Daniela Pittauer, Germany <b>MAN-MADE RADIONUCLIDES IN SEDIMENTS OF THE INDONESIAN THROUGHFLOW</b> <i>Daniela Pittauer, Per Roos, Jixin Qiao, Stephen Tims, Michaela Fröhlich, Helmut W. Fischer</i>
12:00 O1 A-3  Tetsuya Matsunaka, Japan <b>CHANGES IN <sup>129</sup>I/<sup>127</sup>I RATIO OF CRATER LAKE AND VOLCANIC ACTIVITY AT ZAO VOLCANO, JAPAN</b> <i>Tetsuya Matsunaka, Kimikazu Sasa, Keisuke Sueki, Tsutomu Takahashi, Masumi Matsumura, Akio Goto, Takahiro Watanabe, Noriyoshi Tsuchiya, Nobuo Hirano, Hiroyuki Matsuzaki</i>	12:15 O1 B-3  Galina Lujaniene, Lithuania <b>APPLICATION OF <sup>137</sup>Cs, <sup>14</sup>C AND Pu ISOTOPES TO TRACE POLLUTANTS IN THE BALTIC SEA</b> <i>Galina Lujaniene, Jonas Mazeika</i>
12:15 O1 A-4  Silke Merchel, Germany <b>LONG-LIVED COSMOGENIC RADIONUCLIDES: DETERMINATION BY ACCELERATOR MASS SPECTROMETRY AND MODEL APPLICATIONS</b> <i>Silke Merchel, Santiago M. Enamorado Baez, Stefan Pavetich, Georg Rugel, Dreams-Users Dreams-Friends</i>	12:30 O1 B-4  Celine Duffa, France <b>CONSEQUENCES ON MARINE ENVIRONMENT OF A HYPOTHETICAL ACCIDENTAL RADIOACTIVE RELEASE IN THE BAY OF TOULON: IMPLEMENTATION OF A MARINE DISPERSION MODEL</b> <i>Christiane Dufresne, Celine Duffa, Vincent Rey</i>
12:30 O1 A-5  Karin Hain, Germany <b>DETECTION OF Np AND Pu IN SEA WATER WITH REGARD TO THE FUKUSHIMA ACCIDENT USING AMS</b> <i>Karin Hain, Boyana Deneva, Thomas Faestermann, Leticia Fimiani, José Manuel Gomez Guzman, Gunther Korschinek, Florian Kortmann, Christoph Lierse V. Gostomski, Peter Ludwig</i>	12:45 O1 B-5  Georgios Eleftheriou, Greece <b>RECENT COASTAL SEDIMENTATIONS RATES FROM NE MEDITERRANEAN USING <sup>210</sup>Pb<sub>EX</sub> AND <sup>137</sup>Cs</b> <i>Georgios Eleftheriou, Christos Tsabaris, Dionisis Patiris, Effrosyni Androulakaki, Filothei Pappa</i>
12:45 O1 A-6  Rafael García-Tenorio, Spain <b>THE IMPACT OF FUKUSHIMA ACCIDENT IN SEVILLA (SPAIN) THROUGH THE ANALYSIS OF I-129 BY ACCELERATOR MASS SPECTROMETRY</b> <i>Jose María López-Gutierrez, Lidia Agulló, Jose Ignacio Peruchena, José Manuel Gómez-Guzmán, Rafael García-Tenorio</i>	

13:00-14:00 Light Lunch

14:00- 15-00  
PS 1  
Poster Session 1 – Coffee  
ROOM: OSTRIA

	<b>SESSION 2A: RADIOANALYTICS</b> ROOM: MAISTROS A CHAIR: X. HOU & G. LUJANIENÉ
<b>15:00</b> <b>O2 A-1</b>	Sheldon Landsberger, USA <b>DISEQUILIBRIUM IN THE URANIUM AND ACTINIUM SERIES IN OIL SCALE SAMPLES</b> <i>Sheldon Landsberger, Michael Yoho, Dimitri Tamalis, Ckeurcheley Leblanc</i>
<b>15:30</b> <b>O2 A-2</b>	Jakub Kaizer, Slovakia <b>ACCELERATOR MASS SPECTROMETRY LABORATORY AT THE COMENIUS UNIVERSITY IN BRATISLAVA: FIRST RESULTS</b> <i>Pavel P. Povinec, Jozef Masarik, Miroslav Jeřkovský, Róbert Breier, Jakub Kaizer, Andrej Kováčik, Ján Pánik, Marta Richtáriková, Alexander Šivo, Jaroslav Staniček, Jakub Zeman, Peter Steier, Alfred Priller</i>
<b>15:45</b> <b>O2 A-3</b>	Hugues Paradis, France <b>MEASUREMENT BY GAMMA RAY SPECTROMETRY IN GAMMA-GAMMA COINCIDENCE</b> <i>Hugues Paradis, Anne de Vismes, Rodolfo Gurriaran, Xavier Cagnat, Fabrice Piquemal</i>
<b>16:00</b> <b>O2 A-4</b>	Kamila Johnova, Czech Republic <b>MONTE CARLO SIMULATION OF PROBE RESPONSE TO CALIBRATION MEASUREMENTS, STRAZ POD RALSKEM CALIBRATION FACILITY, CZECH REPUBLIC</b> <i>Kamila Johnova, Lenka Thinova, Radek Cerny</i>
<b>16:15</b> <b>O2 A-5</b>	Daniela Ene, Sweden <b>EVALUATION OF THE ENVIRONMENTAL IMPACT OF THE EUROPEAN SPALLATION SOURCE FACILITY</b> <i>Daniela Ene</i>

<b>16:30 -17:00</b>	Coffee Break
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<b>17:00</b> <b>O2 A-6</b>	Patrick Kessler, Germnay <b>A NEW GENERATION OF DETECTORS FOR ENVIRONMENTAL RADIATION MONITORING</b> <i>Patrick Kessler, Harald Dombrowski, Stefan Neumaier</i>
<b>17: 15</b> <b>O2 A-7</b>	Evelyn Krawczyk-Bärsch, Germany <b>USE OF SPECTROSCOPIC AND MICROSCOPIC METHODS TO REVEAL THE EVIDENCE OF U(VI) SORPTION ON ACIDOVORAX FACILIS ISOLATED FROM SUBSURFACE ENVIRONMENTS</b> <i>Evelyn Krawczyk-Bärsch, Ulrike Gerber, Robin Steudtner, Thuro Arnold</i>
<b>17:30</b> <b>O2 A-8</b>	Sara R. H. Vanderheyden, Belgium <b>COMPARISON OF ADSORPTION PROPERTIES OF C<sub>6</sub> ON DIFFERENT TYPES OF ACTIVATED CARBON</b> <i>Sara R. H. Vanderheyden, Raf Van Ammel, Katarzyna Sobiech-Matura, Kenny Vanreppelen, Sonja Schreurs, Wouter Schroeyers, Jan Yperman, Robert Carlee</i>
<b>17:45</b> <b>O2 A-9</b>	Vladimir Sladkov, France <b>AFFINITY CAPILLARY ELECTROPHORESIS IN STUDYING THE COMPLEX EQUILIBRIUMS OF RADIONUCLIDES IN AQUEOUS SOLUTION</b> <i>Vladimir Sladkov</i>
<b>18:00</b> <b>O2 A-10</b>	K. Schomäcker, Germany <b>RETENTION EFFICACY AND RELEASE OF RADIOIODINE IN FUMEHOODS</b> <i>Klaus Schomaecker, Thomas Fischer, Beate Zimmermanns, Jörg Bregulla, Ferdinand Sudbrock, Alexander Drzezga</i>

<b>18:15</b>	Honorary Session for retired Greek Professors pioneers in the field of Environmental Radioactivity in Greece CHAIR: A. IOANNIDOU ROOM: MAISTROS A
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	<b>SESSION 2B: AQUATIC</b> ROOM: SIROCCO CHAIR: CH. TSABARIS & A. CLOUVAS
<b>15:00</b> <b>O2 B-1</b>	Sang-Han Lee, Korea <b>DISTRIBUTION OF RADIONUCLIDES IN THE SEDIMENT OF THE SOUTHERN MEDITERRANEAN SEA</b> <i>Sang-Han Lee, Pavel Povinec, Mai Pham, Beat Gasser and Jean-Luc Pontis</i>
<b>15:15</b> <b>O2 B-2</b>	Heleny Florou, Greece <b>RADIATION DOSE ASSESSMENT OF <sup>137</sup>Cs IN MARINE FISH OF DIFFERENT HABITATS IN THE AEGEAN SEA – GREECE</b> <i>Heleny Florou, Georgia Trabidou, Nikolaos Evangeliou, Georgios Kuburas</i>
<b>15:30</b> <b>O2 B-3</b>	Önder Kılıç Gülşah Kalaycı, Turkey <b>SEASONAL AND SPATIAL DISTRIBUTIONS AND FLUXES OF <sup>137</sup>Cs, <sup>226</sup>Ra, <sup>228</sup>Ra, AND <sup>40</sup>K IN THE GOLDEN HORN ESTUARY, ISTANBUL</b> <i>Önder Kılıç Gülşah Kalaycı, , Murat Belivermiş, Narin Sezer, Furkan Gözel</i>
<b>15:45</b> <b>O2 B-4</b>	Helmut W. Fischer, Germany <b>MEDICAL I-131 IN GERMAN RIVERS</b> <i>Helmut W. Fischer, Susanne Ulbrich, Maria-Evangelia Souti, Rainer Gellermann</i>
<b>16:00</b> <b>O2 B-5</b>	Ushko Natalia, Poland <b>CHEMICAL AND ISOTOPE COMPOSITION IN SELECTED BELARUS MINERAL AND THERAPEUTIC WATERS</b> <i>Ushko Natalia, Nguyen Dinh Chau</i>
<b>16:15</b> <b>O2 B-6</b>	Hedvig Simon, Romania <b>INSIGHT ON THE GEOCHRONOLOGY AND HEAVY METAL CONCENTRATION OF THE IACOB LAKE, DANUBE DELTA, ROMANIA</b> <i>Hedvig Simon, Robert-Csaba Begy, Luminita Preoteasa</i>

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<b>17:00</b> <b>O2 B-7</b>	Juan Mantero, Sweden <b>LEVELS OF NATURAL RADIONUCLIDES IN PIT LAKES FROM COUNTRIES IN AFRICA AND EUROPE</b> <i>Juan Mantero, Guillermo Manjon, , Rimon Thomas, Ignacio Vioque, Inmaculada Diaz-Frances, Jose Antonio Galvan, Abdesalam Absi, Mouloud Lehitane, Said Chakiri, Zhora Bejjaji, Fatima El Hmid, Mats Isaksson, Eva Forssell-Aronsson, Elis Holm and Rafael Garcia-Tenorio</i>
<b>17: 15</b> <b>O2 B-8</b>	S. Sinan Keskin, Turkey <b>APPROXIMATE DATING OF A SEDIMENT CORE OBTAINED FROM TUZ LAKE IN TURKEY USING Cs-137 ISOTOPIC MEASUREMENTS</b> <i>S. Sinan Keskin and Ozlem Guner</i>
<b>17: 30</b> <b>O2 B-9</b>	José Antonio Corcho Alvarado, Switzerland <b>VARIABILITY IN THE URANIUM ISOTOPIC RATIOS IN THE VICINITY OF FORMER URANIUM MINING AND MILLING SITES</b> <i>José Antonio Corcho Alvarado, Beatrice Balsiger, Mario Burger, Stefan Röllin, Hans Sahli, Alfred Jakob</i>
<b>17:45</b> <b>O2 B-10</b>	Marilyne Stuart, Canada <b>ARE DRINKING WATER GUIDELINES FOR TRITIUM ADEQUATE TO PROTECT AQUATIC BIOTA?</b> <i>Marilyne Stuart, Amy Festarini, Sang-Bog Kim, Carmen Shultz, Heather Ikert, Krista Schleicher, Stephanie Walsh, Cecile Dubois, Beatrice Gagnaire</i>
<b>18:00</b> <b>O2 B-11</b>	Hasan Baltas, Turkey <b>RADIONUCLIDE CONCENTRATIONS IN MUSSELS (MYTILUS GALLOPROVINCIALIS) COLLECTED FROM THE EASTERN BLACK SEA REGION OF TURKEY IN 2014</b> <i>Hasan Baltas, Erkan Kiris, Goktug Dalgic, Esra Yilmaz Bayrak, Gokhan Apaydin, Ugur Cevik</i>

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08:30-09:00  
PS 1

Poster Session 1  
ROOM: OSTRIA

09:00-10:30 PL2	PLENARY SESSION 2 ROOM: MAISTROS A, CHAIR: O. MASSON, P. VOJTYLA
09:00 PL2-1	Katsumi Hirose, Japan FUKUSHIMA ACCIDENT AND ENVIRONMENTAL RADIOACTIVITY Katsumi Hirose
09:30 PL2-2	Michio Aoyama, Japan 134Cs AND 137Cs IN THE NORTH PACIFIC OCEAN DERIVED FROM THE TEPCO FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT ACCIDENT IN MARCH 2011 Michio Aoyama, Yasunori Hamajima, Mikael Hult, Mitsuo Uematsu, Eitaro Oka, Daisuke Tsumune, Yuichiro Kumamoto
10:00 PL2-3	Mark Baskaran, USA APPLICATIONS OF RADON AS A TRACER IN ATMOSPHERIC STUDIES: LESSONS LEARNED AND FUTURE CHALLENGES Mark Baskaran
10:30-11:00	Coffee Break

PARALLEL SESSIONS 3 11:00-13:00

	SESSION 3A: FUKUSHIMA ROOM: MAISTROS A CHAIR: K. HIROSE & M. AOYAMA
11:00 O3 A-1	Daisuke Tsumune, Japan FOUR-YEARS REGIONALSCALE SIMULATION OF 137Cs RADIOACTIVITY IN THE OCEAN FOLLOWING THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT Daisuke Tsumune, Michio Aoyama, Katsumi Hirose, Takaki Tsubono, Kazuhiro Misumi Yutaka Tateda
11:15 O3 A-2	Sabine Charmasson, France 137Cs AND TRITIUM CONCENTRATIONS IN SEAWATER OFF THE FUKUSHIMA PREFECTURE: RESULTS FROM THE SOSO 5 RIVERS CRUISE (OCTOBER 2014) Michio Aoyama, Sabine Charmasson, Yasunori Hamajina, Herve Thebault, Mireille Arnaud, Celine Duffa, Franck Giner
11:30 O3 A-3	Yutaka Tateda, Japan BIOKINETICS OF RADIOCESIUM DEPURATION IN MARINE FISH INHABITING THE VICINITY OF THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT Yutaka Tateda, Daisuke Tsumune, Kazuhiro Misumi
11:45 O3 A-4	Kazuhiro Nakashoji, Japan DEVELOPMENT OF 236U-AMS IN MALT Kazuhiro Nakashoji, Hiroyuki Matsuzaki, Yasuto Miyake, Aya Sakaguchi
12:00 O3 A-5	Vasyl Yoschenko, Japan RADIOCESIUM DISTRIBUTION AND FLUXES IN THE TYPICAL CRYPTOMERIA JAPONICA FOREST AT THE LATE STAGE AFTER THE ACCIDENT AT FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT Vasyl Yoschenko, Tsugiko Takase, Sergiy Kivva, Alexei Konoplev, Kenji Nanba, Yuichi Onda, Mark Zheleznyak, Natsumi Sato, Koji Keitoku
12:15 O3 A-6	Alexei Konoplev, Japan RADIOCESIUM SOLID-LIQUID DISTRIBUTION, WASH-OFF AND MIGRATION WITHIN CONTAMINATED CATCHMENTS AFTER THE ACCIDENT AT FUKUSHIMA DAIICHI NUCLEAR POWER PLANT Alexei Konoplev, Valentin Golosov, Sergei Kivva, Kenji Nanba, Kiyoshi Omine, Yuichi Onda, Tsugiko Takase, Yoshifumi Wakiyama, Vasyl Yoschenko, Mark Zheleznyak
12:30 O3 A-7	Airi Mori, Japan ASSESSMENT OF RESIDUAL DOSES TO THE POPULATION AFTER THE DECONTAMINATION WORKS IN FUKUSHIMA PREFECTURE Airi Mori, Shogo Takahara, Azusa Ishizaki, Masashi Iijima, Yukihisa Sanada, Masahiro Munaka

13:00-14:00

Light Lunch

	SESSION 3B: NORM ROOM: SIROCCO CHAIR: S. LANDSBERGER & M. BASKARAN
11:00 O3 B-1	Rafael Garcia-Tenorio, Spain RADIOACTIVE CHARACTERIZATION OF RESIDUES FROM AN OIL INDUSTRY IN GHANA David O. Kpeglo, Juan Mantero, E.O. Darko, G. Emi-Reynolds, A. Faanu, Guillermo Manjon, Ignacio Vioque, E.H.K. Akaho, Rafael Garcia-Tenorio
11:30 O3 B-2	Silvia Pérez Moreno, Spain MOBILITY OF RADIUM IN PHOSPHOGYPSUM WASTE GENERATED BY THE WET SULPHURIC PATHWAY. Silvia Pérez Moreno, Elisa Rodriguez, Manuel J. Gázquez, Jose Luis Guerrero, Fernando Mosqueda, Juan Pedro Bolívar
11:45 O3 B-3	Fernando Mosqueda Peña, Spain OCCUPATIONAL EXPOSURES IN TWO INDUSTRIAL PLANTS DEVOTED TO THE PRODUCTION OF AMMONIUM PHOSPHATE FERTILIZERS Fernando Mosqueda Peña, Juan Pedro Bolívar Raya, Manuel Jesús Gázquez González, Silvia Pérez Moreno, Federico Vaca Galán, Rafael García-Tenorio García-Balmaseda
12:00 O3 B-4	Filothei Pappa, Greece NORM MEASUREMENTS AT THE BEACH SAND AND COASTAL SEDIMENTS NEAR A MINING AREA, IERISSOS GULF, GREECE Filothei Pappa, Christos Tsabaris, Dionisis Patiris, Helen Kaberi, Christina Zeri, Eyfrosini Androulakai, Georgios Eleftheriou, Michael Kokkoris, Roza Zanni-Vlastou, Alexandra Ioannidou
12:15 O3 B-5	Petra Planinšek, Slovenia TRANSFER OF Ra-266 FROM SOIL CONTAMINATED WITH U-MILL TAILING TO RADISH (RAPHANUSSATIVUS L.) AND SAVOY (BRASSICA OLERANCEA L. VAR. SABAUDA) Petra Planinšek, Borut Smodiš, Ljudmila Benedik
12:30 O3 B-6	Maria Nikolaki, Greece NATURAL RADIOACTIVITY OF BUILDING MATERIALS IN GREECE Maria Nikolaki, Georgios Takoudis, Constantinos Potiriadis
12:45 O3 B-7	Juan Mantero, Spain CHARACTERIZATION OF SCALES FROM A NORM (TiO2) INDUSTRY Juan Mantero, Ignacio Vioque, Manuel Gazquez, Juan Pedro Bolivar, Rafael Garcia-Tenorio

14:00-15:00 Poster Session 2 – Coffee  
PS 2 ROOM: MAISTROS A  
Ro 5 Ro5 meeting  
ROOM: MAISTROS C

PARALLEL SESSIONS 4 15:00-18:15

SESSION 4A: SOIL

ROOM: MAISTROS A  
CHAIR: H.W. FISCHER & M. KRMAR

- 15:00  
O4 A-1

Peter Bossew, Germany

**SIMPLE ANALYTICAL MODELS OF RADIONUCLIDE TRANSPORT IN TOPSOIL**

*Peter Bossew, Sarat Sahoo, Suchismita Mishra*
- 15:30  
O4 A-2

Hirose Katsumi, Japan

**PLUTONIUM AND URANIUM ISOTOPES IN MONGOLIA SURFACE SOILS**

*Hirose Katsumi, Yoshikazu Kikawada, Yasuhito Igarashi*
- 15:45  
O4 A-3

Sarata Kumar Sahoo, Japan

**MASS SPECTROMETRIC MEASUREMENT OF URANIUM ISOTOPE RATIOS IN NUCLEAR ACCIDENT CONTAMINATED SOIL SAMPLES**

*Asako Takamasa, Sarata Kumar Sahoo, Shun'ichi Nakai, Hideki Arae*
- 16:00  
O4 A-4

Caroline Licour, Belgium

**<sup>226</sup>Ra IN BELGIAN SOILS: SYNTHESIS OF STUDENT WORKS AT ISIB**

*Caroline Licour*

SESSION 4B: NORM/ Radon

ROOM: SIROCCO  
CHAIR: R. GARCIA-TENORIO & A. VARGAS

- 15:00  
O4 B-8

Hyuncheol Kim, Korea

**VALIDATION OF THE METHOD OF DETERMINATION OF <sup>226</sup>Ra IN NORM BY LSC: COMPARISON WITH ICP-MS AND GAMMA SPECTROMETER METHOD**

*Hyuncheol Kim, Young-Yong Ji, Jong Myung Lim, Young Gun Ko, Kun Ho Chung, Yoonhee Jung*
- 15:15  
O4 B-9

Hiromi Yamazawa, Japan

**OBSERVATION AND MODELLING OF TEMPORAL VARIATION IN RADON DECAY PRODUCT CONCENTRATION IN RAIN WATER**

*Hiromi Yamazawa, Keisuke Sakata, Heng Liu, Shigekazu Hirao, Jun Moriizumi*
- 15:30  
O4 B-10

Panagiota Semertzidou, UK

**A DYNAMICAL APPROACH IN THE DISTRIBUTION PATTERNS OF NATURAL RADIONUCLIDES DECAYING FROM RADON-222 IN THE ATMOSPHERE**

*Panagiota Semertzidou, Gayane Piliposyan, Peter Appleby*
- 15:45  
O4 B-11

Giorgia Cinelli, Italy

**MAPPING OF THE DOSE RATE FROM GROUND NATURAL GAMMA RADIATION IN THE EUGANEAN HILLS: MEASURED DOSE AND BEDROCK CONCENTRATION OF RADIONUCLIDES**

*Domiziano Mostacci, Giorgia Cinelli, Laura Tositti, Raffaele Sassi and Claudio Mazzoli*
- 16:00  
O4 B-12

Georg. Bátor, Hungary

**DISTRIBUTION OF RADIOACTIVE MATERIALS IN THE SAND SAMPLES OF COASTAL AREAS ALONG THE AEGEAN SEA (GREECE) AND DOSE ASSESSMENT OF NATURAL RADIATION**

*Amin Shahrokhi, G. Bátor, Jónás Jácint, Tibor Kovács*

16:15- 16:45 Coffee Break

- 16:45  
O4 A-5

Laura Arata, Switzerland

**THE <sup>137</sup>Cs REPEATED SAMPLING APPROACH TO DERIVE SOIL REDISTRIBUTION RATES AND VALIDATE REFERENCE SITES IN ALPINE GRASSLANDS**

*Laura Arata, Carmelo La Spada, Katrin Meusburger, Markus Zehringner, Lionel Mabit, Christine Alewell*
- 17:00  
O4 A-6

Maria Angelica Vergara Wasserman, Brazil

**CONCEPTUAL MODEL TO CATEGORIZE AND MAP VULNERABILITY OF SOILS TO <sup>137</sup>Cs CONTAMINATION**

*Maria Angelica Vergara Wasserman, Pericles Picanço Jr, Daniel Vidal Pérez, Fernanda Leite Silva, Celso Marcelo Lapa, Bary Nóbrega, Elaine Rua R. Rochedo, Ralph Santos-Oliveira*
- 17:15  
O4 A-7

Eleni Siasou, U.K.

**AN ANALYSIS OF INTRA-SPECIES AND INTER-SPECIES DIFFERENCES IN UPTAKE OF RADIOIODINE FROM SOIL BY FLOWERING PLANTS**

*Eleni Siasou, Janine Wilkins, Neil Willey*
- 17:30  
O4 A-8

Dimitrios Kosmidis, U.K.

**INVESTIGATING THE ROLE OF NATURAL ORGANIC MATTER (NOM) IN THE MIGRATION OF KEY RADIONUCLIDES IN SOUTH TERRAS, CORNWALL, ENGLAND**

*Dimitrios Kosmidis, Nick Evans, Monica Felipe - Sotelo, Joanna Kulaszewska and David Read*
- 17:45  
O4 A-9

Gergo Bátor, Hungary

**RADIOCARBON MONITORING IN THE VICINITY OF THE HUNGARIAN NPP**

*Gergő Bátor, András Bednár, Tibor Kovács*
- 18:00  
O4 A-10

Sofija Forkapic, Serbia

**ASSESSMENT OF SOIL EROSION RATE BASED ON RADIONUCLIDE ACTIVITY CONCENTRATIONS**

*Sofija Forkapic, Kristina Bikit, Dusan Mrdja and Istvan Bikit*

- 16:45  
O4 B-13

Kristina Bikit, Serbia

**RADIOACTIVITY IN SPECIAL NATURE RESERVE “ZASAVICA”**

*Dusan Mrdja, Kristina Bikit, Sofija Forkapić, Istvan Bikit, Marko Cvijanovic*
- 17:00  
O4 B-14

Naglaa Fahmi, Egypt

**RADON CONCENTRATION MEASUREMENTS IN GATTAR URANIUM PROSPECT, NORTHERN EASTERN DESERT, EGYPT**

*Naglaa Fahmi*
- 17:15  
O4 B-15

Fani Papageorgiou, Greece

**ENVIRONMENTAL AND RADIOLOGICAL RISK ASSESSMENT OF THE SCHISTOS PHOSPHOGYPSUM WASTE REPOSITORY**

*Fani Papageorgiou, Athanasios Godelitsas, Nikolaos Voulgaris, Theodoros Mertzimekis, Stelios Xanthos and Georgios Katsantonis*
- 17:30  
O4 B-16

Bulgasem El-Fawaris, Libya

**REMOVAL OF TE-NORM SCALES FROM GAS-OIL-WATER SEPARATORS IN LIBYAN OIL FIELDS**

*Bulgasem El-Fawaris, Hans Doerfel*

08:30-09:00  
PS 2

Poster Session 2  
ROOM: OSTRIA

09:00-10:30 PL3	PLENARY SESSION 3 ROOM: MAISTROS A, CHAIR: A.J.T. JULL & P. STEIER
09:00-09:30 PL3-1	William Kieser, Canada NEW APPLICATIONS IN ACCELERATOR MASS SPECTROMETRY USING DEVELOPMENTS OF ION SOURCE AND ION-GAS REACTION CELL TECHNOLOGY <i>William Kieser, Xiaolei Zhao, John Eliades, Chris Charles, Cole MacDonald, Jack Cornett, Ted Litherland</i>
09:30-10:00 PL3-2	Mihály Molnár, Hungary HIGH PRECISION <sup>14</sup> C ANALYSES BY SMALL AMS - APPLICATIONS IN NUCLEAR ENVIRONMENTAL PROTECTION <i>Mihály Molnár, László Rinyu, Róbert Janovics, István Major, Anita Molnár, Mihály Veres, A.J. Timothy Jull</i>
10:00-10:30 PL3-3	Xiaolin Hou, Denmark MASS SPECTROMETRY FOR DETERMINATION OF ENVIRONMENTAL RADIONUCLIDES <i>Xiaolin Hou</i>

10:30-11:00

Coffee Break

PARALLEL SESSIONS 5 11:00-12:30

	SESSION 5A: GROUND WATER ROOM: MAISTROS A CHAIR: D. TSUMUNE & Y. TATEDA
11:00 O5 A-1	Daniel Bonotto, Brazi; THE DISSOLVED URANIUM CONCENTRATION AND <sup>234</sup> U/ <sup>238</sup> U ACTIVITY RATIO IN MINERAL WATERS OF SOUTHEASTERN BRAZIL <i>Daniel Bonotto</i>
11:15 O5 A-2	Konstantina Kehagia, Greece NATURAL RADIONUCLIDES IN THERMAL MINERAL SPRINGS IN EDIPSOS GREECE <i>Konstantina Kehagia, Sotirios Bratakos, Dimitrios Xarchoulakos, Konstantinos Potiriadis</i>
11:30 O5 A-3	Sergey Subbotin, Kazakhstan CHARACTER AND LEVELS OF RADIOACTIVE CONTAMINATION OF UNDERGROUND WATER AT THE SEMIPALATINSK TEST SITE <i>Sergey Subbotin, Sergey Lukashenko, Elena Novikova</i>
11:45 O5 A-4	Alfatih Osman, Germany TRITIUM ACTIVITY LEVELS IN NATURAL WATERS AROUND HANNOVER, NORTHERN GERMANY <i>Alfatih Osman, Stefan Bister, Beate Riebe, Clemens Walther</i>
12:00 O5 A-5	Oxana Lyakhova, Kazakhstan ASSESSMENT OF CHARACTER OF TRITIUM DISTRIBUTION IN AIR OF NUCLEAR TESTING SITES <i>Oxana Lyakhova, Sergey Lukashenko and Lubov Timonova</i>
12:15 O5 A-6	Wagner de Souza Pereira, Brazil NATURAL RADIONUCLIDES IN SURFACE WATER IN A NATURALLY OCCURRING RADIOACTIVE MATERIAL AREA IN BRAZIL <i>Wagner de Souza Pereira, Alphonse Kelecom, Ademir Xavier Da Silva</i>

12:30

Vergina & Winery Conference Tour

	SESSION 5B: RADIOECOLOGY I ROOM: SIROCCO CHAIR: P. BOSSEW & A. BOLSUNOVSKY
11:00 O5 B-1	Antonia Camacho, Spain LONG-TERM RADIOLOGICAL CHARACTERIZATION OF WATERS AT ONE CATALAN DRINKING WATER TREATMENT PLANT <i>Antonia Camacho, Dani Mulas, Isabel Serrano, Sonia Blazquez, R. CardeÑoso, Ricard Devesa Maria Amor Duch</i>
11:15 O5 B-2	Alexandros Clouvas, Greece TIME SERIES ANALYSIS OF LONG TERM (1988-2015) GAMMA RADIATION MEASUREMENTS IN THESSALONIKI, GREECE. <i>Alexandros Clouvas, Fokion Leontaris, Stelios Xanthos, Leontios Hadjileontiadis</i>
11:30 O5 B-3	Dawn Wellman, USA PHOENIX: A WEB-BASED PLATFORM FOR ACCESSING AND EVALUATING HANFORD SITE GROUNDWATER CONTAMINANT DATA <i>M.B. Triplett, T.E. Seiple, D.J. Watson, Dawn Wellman</i>
11:45 O5 B-4	Emily Caffrey, USA TESTING VOXEL MODELS: VOXELIZED VS. ELLIPSOIDAL RABBIT MODELS FOR USE IN RADIOLOGICAL DOSE RATE CALCULATIONS <i>Emily Caffrey, Mathew Johansen, Kathryn Higley</i>
12:00 O5 B-5	Elisabeth Leclerc, France IMPLEMENTING A GEOLOGICAL DISPOSAL OF RADIOACTIVE WASTE IN FRANCE – STRATEGY FOR RADIOLOGICAL ZERO-STATE DEFINITION OF THE ENVIRONMENTAL IMPACT ASSESSMENT (EIA) <i>Elisabeth Leclerc, Philippe Calmon</i>
12:15 O5 B-6	Nathalie Vanhoudt, Belgium SELECTION OF PHYTO/PHYCO-REMEDATION SYSTEMS FOR RADIONUCLIDE DECONTAMINATION <i>Nathalie Vanhoudt, Paul Janssen, Natalie Leys, Hildegarde Vandenhove</i>

<b>09:00-10:30</b> <b>PL4</b>	<b>PLENARY SESSION 4</b>  <b>ROOM: MAISTROS A, CHAIR: W. PLASTINO &amp; J. MIETELSKI</b>
<b>09:00-09:30</b> <b>PL4-1</b>	Olivier Masson, France <b>EUROPEAN-SCALE INVESTIGATIONS ON RADIONUCLIDES IN THE ATMOSPHERE: KNOWLEDGE IMPROVEMENTS CONSECUTIVE TO THE FUKUSHIMA ACCIDENT</b> <i>Olivier Masson, Jackie Tav, Anne De Vismes-Ott, Xavier Cagnat, Pascal Paulat, Rodolfo Gurriaran, Christelle Antonelli, Guillaume Manifica</i>
<b>09:30-10:00</b> <b>PL4-2</b>	Mai Khanh Pham, Monaco <b>PRODUCTION OF IAEA CERTIFIED REFERENCE MATERIALS FOR RADIONUCLIDES IN MARINE ENVIRONMENT STUDIES</b> <i>Mai Khanh Pham, Arend Victor Harms, Iolanda Osvath</i>
<b>10:00-10:30</b> <b>PL4-3</b>	Pavol Vojtyla, Switzerland <b>RADIOLOGICAL ENVIRONMENTAL ASPECTS OF HIGH-POWER ACCELERATOR FACILITIES: CAN THEY BE SHOWSTOPPERS?</b> <i>Pavol Vojtyla</i>

<b>10:30-11:00</b>	Coffee Break
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	<b>PARALLEL SESSIONS 6 11:00-13:00</b>
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	<b>SESSION 6A: ATMOSPHERE</b> ROOM: MAISTROS A CHAIR: M.K. PHAM, S.-H. LEE		<b>SESSION 6B: RADIOECOLOGY II</b> ROOM: SIROCCO CHAIR: K. KEHAGIA & F. GROPPi
<b>11:00</b> <b>O6 A-1</b>	Anne de Vismes Ott, France <b>RADIOLOGICAL SURVEILLANCE OF AEROSOL AND FALLOUT IN FRANCE: LAST IMPROVEMENTS IN TERMS OF SAMPLING AND MEASUREMENT</b> <i>Anne de Vismes Ott, Olivier Masson, Xavier Cagnat, Rodolfo Gurriaran</i>	<b>11:00</b> <b>O6 B-1</b>	Alexander Bolsunovsky, Russia <b>RADIOACTIVE PARTICLES IN THE FLOODPLAIN OF THE YENISEI RIVER</b> <i>Alexander Bolsunovsky, Dmitry Dementyev, Tatiana Zotina, Mikhail Melgunov</i>
<b>11:15</b> <b>O6 A-2</b>	Wolfango Plastino, Italy <b>DETERMINATION OF THE GLOBAL COVERAGE OF THE CTBTO IMS XENON-133 COMPONENT FOR THE DETECTION OF NUCLEAR EXPLOSIONS</b> <i>Wolfango Plastino, Michael Schöppner</i>	<b>11:15</b> <b>O6 B-2</b>	Volker Hormann, Germany <b>MODELLING DISTRIBUTION AND TRANSPORT OF MEDICAL I-131 IN A WASTEWATER TREATMENT PLANT</b> <i>Volker Hormann, Helmut W. Fischer</i>
<b>11:30</b> <b>O6 A-3</b>	Philipp Steinmann, Switzerland <b>ARE COSMOGENIC Be-7 AND Na-22 OR ARTIFICIAL ISOTOPES (LEGACY OF ATMOSPHERIC NUCLEAR WEAPON TESTS) TRACERS OF STRATOSPHERIC INPUTS IN GROUNDLEVEL AIR?</b> <i>Philipp Steinmann, Ulla Heikkilä</i>	<b>11:30</b> <b>O6 B-3</b>	Árpád Bihari, Hungary <b>ENVIRONMENTAL RADIOACTIVITY AND SITE-SPECIFIC CONCENTRATION RATIOS OF NON-HUMAN BIOTA IN THE VICINITY OF PAKS NPP, HUNGARY</b> <i>Árpád Bihari, Zoltán Dezső, Róbert Janovics, Andrea Czébely, Judit Orsovszki, Mihály Veres</i>
<b>11:45</b> <b>O6 A-4</b>	Maria Evangelia Souti, Germany <b>Be-7 IN VARIOUS ENVIRONMENTAL MEDIA IN THE BREMEN REGION</b> <i>Maria Evangelia Souti, Helmut Fischer, Prince Benjamin Amoo</i>	<b>11:45</b> <b>O6 B-4</b>	Ferdinand Sudbrock, Germany <b>EXHALATION OF <sup>131</sup>I AFTER RADIOIODINE THERAPY - DOSIMETRIC CONSIDERATIONS BASED ON MEASUREMENTS IN EXHALED AIR</b> <i>Klaus Schomaecker, Ferdinand Sudbrock, homas Fischer, Beate Zimmermanns, Alex.Drzezga</i>
<b>12:00</b> <b>O6 A-5</b>	Galina Lujanieni, Lithuania <b>SOURCES OF <sup>137</sup>Cs, <sup>241</sup>Am AND Pu ISOTOPES IN THE ATMOSPHERE</b> <i>Galina Lujanieni, Steigvile Byčenkieni</i>	<b>12:00</b> <b>O6 B-5</b>	Peter Bossew, Germany <b>ESTIMATION OF THE GEOGENIC RADON POTENTIAL AND RADON PRONE AREAS FROM GEOCHEMICAL QUANTITIES AND AMBIENT DOSE RATE</b> <i>Peter Bossew, Giorgia Cinelli, Tore Tollefsen, Valeria Gruber,Marc De Cort</i>
<b>12:15</b> <b>O6 A-6</b>	Ekaterini Dalaka, Greece <b>SEASONAL VARIABILITY OF <sup>7</sup>Be AMAD &amp; DEPENDENCY ON ATMOSPHERIC CONDITIONS IN ATHENS</b> <i>Ekaterini Dalaka, Marios Anagnostakis, Konstantinos Eleftheriadis</i>	<b>12:15</b> <b>O6 B-6</b>	Alua Kabdyrakova, Kazakhstan <b>RADIONUCLIDES DISTRIBUTION IN SOIL GRAIN-SIZE FRACTIONS IN PLACES OF NUCLEAR TESTS AT SEMIPALATINSK TEST SITE</b> <i>Alua Kabdyrakova, Assiya Kunduzbaeva and Sergey Lukashenko</i>
<b>12:30</b> <b>O6 A-7</b>	Jerzy W. Mietelski, Poland <b>GAMMA EMITTERS IN ATMOSPHERIC PRECIPITATION IN KRAKOW (SOUTHERN POLAND) SINCE 2005</b> <i>Jerzy W. Mietelski, Ewa Tomankiewicz, Kamil Brudecki, Pawel Janowski, Ewa Nalichowska Renata Kierepko</i>	<b>12:30</b> <b>O6 B-7</b>	Assiya Kunduzbaeva, Kazakhstan <b>RESEARCH ON SPECIES OF ARTIFICIAL RADIONUCLIDES <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>239+240</sup>Pu AND <sup>241</sup>Am IN SOILS OF CONDITIONALLY «BACKGROUND» TERRITORIES OF SEMIPALATINSK TEST SITE</b> <i>Assiya Kunduzbaeva, Alua Kabdyrakova and Sergey Lukashenko</i>
<b>12:45</b> <b>O6 A-8</b>	A. Kováčik, Slovakia <b>SOURCES OF RECENT RADIONUCLIDE VARIATIONS IN THE ATMOSPHERE</b> <i>P. P. Povinec, K. Hirose, Y. Igarashi, A. Ioannidou, M. Jeskovsky, A. Kováčik, G. Lujanienė, J. W. Mietelski, M. K. Pham, I. Šykora, J. Bartok, I. Bartokova</i>	<b>12:45</b> <b>O6 B-8</b>	Abubakar Bajoga, U.K. <b>SOIL RADIOACTIVITY LEVELS AND RADIOLOGICAL RISK EVALUATION ACROSS THE STATE OF KUWAIT</b> <i>Abubakar Bajoga, Naser Alazemi, Hassan Shams, Patrick Regan, David Bradley</i>

<b>13:00-14:00</b>	Lunch
<b>14:00-15:00</b> <b>PS 3</b>	<b>Poster Session 3 – Coffee</b> ROOM: OSTRIA



PARALLEL SESSIONS 7 15:00-18:00	
	<b>SESSION 7A: ATMOSPHERE</b> ROOM: MAISTROS A CHAIR: M. MOLNÁR & I. SVETLIK
<b>15:00</b> <b>07 A-1</b>	Mai Khanh Pham, Monaco <b>AIR MONITORING IN MONACO: STUDY OF SAHARAN DUST TRANSPORT</b> <i>Mai Khanh Pham, Elena Chamizo, Jose Luis Mas Balbuena</i>
<b>15:15</b> <b>07 A-2</b>	Jackie Tav, France <b>DEPOSITION OF RADIONUCLIDES BY CLOUD WATER ON PLANTS</b> <i>Jackie Tav, Olivier Masson, Frédéric Burnet, Pascal Paulat, Thierry Bourrianne, Anne De Vismes, Karine Sellegri</i>
<b>15:30</b> <b>07 A-3</b>	Rafael Correa, Chile <b>DETECTION OF Cs-137 IN THE ANDES CORDILLERA AT LATITUDE 33°56' SOUTH.</b> <i>José Morales, Rafael Correa, Pedro Miranda, Pablo Ortiz, Eduardo Mera, Sofia Camilla, Javier Wachter, Osvaldo Piñones</i>
<b>15:45</b> <b>07 A-4</b>	Miguel Angel Hernández-Ceballos, Italy <b>SEASONALITY OF <sup>7</sup>Be CONCENTRATIONS IN EUROPE AND INFLUENCE OF TROPOPAUSE HEIGHT</b> <i>Miguel Angel Hernández-Ceballos, Erika Brattich, Giorgia Cinelli</i>
<b>16:00</b> <b>07 A-5</b>	Luca De Felice, Italy <b>FEASIBILITY STUDY ON AUTOMATIC EVALUATION OF ANOMALOUS RADIOACTIVITY LEVELS USING INTAMAP INTERPOLATION SERVICE WITHIN EURDEP NETWORK</b> <i>Luca De Felice, Miguel Angel Hernández-Ceballos, Montserrat Marin Ferrer, Konstantins Bogucarskis, Giorgia Cinelli, Marc de Cort, Erika Brattich</i>
<b>16:15-16:45</b>	Coffee Break

<b>16:45</b> <b>07 A-6</b>	Miodrag Krmar, Serbia <b><sup>7</sup>Be AND <sup>210</sup>Pb ATMOSPHERIC DEPOSITION MEASURED BY A MOSS TECHNIQUE AND ITS DEPENDENCE ON CUMULATIVE PRECIPITATION</b> <i>Miodrag Krmar, Dragutin Mihailovic, Ines Pap, Ilija Arsenic</i>
<b>17:00</b> <b>07 A-7</b>	Erika Brattich, Italy <b>APPLICATIONS OF ENVIRONMENTAL RADIONUCLIDES TO ATMOSPHERIC STUDIES AT THE WMO-GAW STATION OF MT. CIMONE</b> <i>Erika Brattich, Laura Tositti, José Antonio Garcia Orza, Hongyu Liu, Miguel Angel Hernandez Ceballos</i>
<b>17:15</b> <b>07 A-8</b>	Kerstin Hürkamp, Germany <b>RADIONUCLIDE DEPOSITION WITH SNOW ON MT. ZUGSPITZE, GERMANY: WASHOUT AND RELEASE TO SURFACE WATER</b> <i>Kerstin Hürkamp, Felix Bernauer, Eva Maria Schiestl, Ludwig Ries, Jochen Tschiersch</i>
<b>17:30</b> <b>07 A-9</b>	Taieb Errahmani Djamel, Algeria <b>SEASONAL VARIATIONS OF RADIOACTIVITY IN AEROSOLS AT ALGIERS STATION, ALGERIA</b> <i>Taieb Errahmani Djamel and Noureddine Abdelkader</i>

	<b>SESSION 7B: RADIOECOLOGY III</b> ROOM: SIROCCO CHAIR: C. COSMA & O. LYAKHOVA
<b>15:00</b> <b>07 B-1</b>	Ferdinand Sudbrock, Germany <b>INFLUENCE OF IODINE SUPPLY ON THE RADIATION-INDUCED DNA-FRAGMENTATION</b> <i>Ferdinand Sudbrock, Anika Herrmann, Thomas Fischer, Beate Zimmermanns, Wolfgang Baus, Alexander Drzezga, Klaus Schomaecker</i>
<b>15:15</b> <b>07 B-2</b>	Constantin Cosma, Romania <b>Cs-137 IN OAK BARK SAMPLES IN SOME REGIONS FROM ROMANIA AND SERBIA</b> <i>Constantin Cosma, Zora Zunic, Marian Mihaiu, Branislava Batos, Sabahudin Hadrovic, Dragan Alavantic, Andra Rada Iurian</i>
<b>15:30</b> <b>07 B-3</b>	Caitlin Condon, USA <b>PROGRESS ON A VOXEL PHANTOM MODEL OF A PINE TREE</b> <i>Caitlin Condon, Kathryn Higley</i>
<b>15:45</b> <b>07 B-4</b>	Natalya Larionova, Kazakhstan <b>SEMIPALATINSK TEST SITE (STS). PRESENT STATE AND PROSPECTS</b> <i>Sergey Lukashenko, Natalya Larionova</i>
<b>16:00</b> <b>07 B-5</b>	Vasiliki Kioupi, Greece <b>BIOACCUMULATION OF THE NATURAL RADIONUCLIDES Th-234, Ra-226 AND K-40 AND THE ARTIFICIAL Cs-137 IN THE FRUITBODIES OF BASIDIOMYCETES IN GREECE</b> <i>Vasiliki Kioupi, Heleny Florou, Zacharoula Gonou-Zagou, Evangelia Kapsanaki-Gotsi</i>

<b>16:45</b> <b>07 B-6</b>	Andrey Panitskiy, Kazakhstan <b>RADIONUCLIDES IN BODIES OF WILD ANIMALS OF SEMIPALATINSK TEST SITE</b> <i>Andrey Panitskiy, Sergey Lukashenko, Nuriya Kadyrova</i>
<b>17:00</b> <b>07 B-7</b>	Zhanat Baigazinov, Kazakhstan <b>PARAMETERS OF RADIONUCLIDE TRANSFER TO LIVESTOCK AND POULTRY PRODUCTS IN DISTANT PERIOD AFTER NUCLEAR TESTS AT THE SEMIPALATINSK TEST SITE</b> <i>Zhanat Baigazinov, Sergey Lukashenko, Andrei Panitskiy</i>
<b>17:15</b> <b>07 B-8</b>	Andrej Kovacic, Slovakia <b>STUDY OF AEROSOL <sup>137</sup>Cs ACTIVITIES AROUND NUCLEAR POWER PLANTS IN SLOVAKIA</b> <i>Miroslav Jeskovsky, Pavel Povinec, Ondrej Slávik, and Ivan Sýkora</i>
<b>17:30</b> <b>07 B-9</b>	Lilla Hoffmann, Hungary <b>IMPROVEMENT OF AN ATMOSHPERIC DISPERSION PROGRAM (SINAC)</b> <i>Lilla Hoffmann, Peter Szanto, Barbara Brockhauser, Sandor Deme, Emese Homolya, Tamas Pazmandi</i>
<b>17:45</b> <b>07 B-10</b>	Peter Szanto, Hungary <b>COMPREHENSIVE ANALYSIS OF 6 YEARS OF ENVIRONMENTAL DOSE DATA MEASURED AT THE TELEMETRIC RADIATION MONITORING STATIONS AROUND PAKS NPP</b> <i>Peter Szanto, Laszlo Manga, Istvan Apathy, Sandor Deme, Attila Hirn, Andras Lencses Tamas Pazmandi</i>



08:30-09:00  
PS3  
Poster Session 3  
ROOM: OSTRIA

09:00-10:00  
PL5  
PLENARY SESSION 5  
ROOM: MAISTROS A, CHAIR: F. BRECHIGNAC, C. PAPASTEFANOU

09:00-09:30  
PL5-1  
Christos Tsabaris, Greece  
IN SITU gamma-RAY SPECTROMETRY IN THE DEEP OCEANS  
Christos Tsabaris, Eyfrosini Androulakaki, Dionisis Patiris, Georgios Eleftheriou, Michael Kokkoris, Filothei Pappa, Roza Zanni-Vlastou

09:30-10:00  
PL5-2  
Wolfgang Plastino, Italy  
ENVIRONMENTAL RADIOACTIVITY AND EARTHQUAKE: A POSSIBLE SIGNATURE OR THE REAL TRIGGER?  
Wolfgang Plastino

10:00-10:30  
PL5-3  
A.Chatt, Canada  
DETERMINATION OF PPB LEVELS OF <sup>238</sup>U AND <sup>232</sup>Th WITH LOW EXPANDED UNCERTAINTIES BY PSEUDO-CYCLIC EPITHERMAL INAA USING COMPTON SUPPRESSION GAMMA-RAY SPECTROMETRY  
A.Chatt

10:30- 11:00  
Coffee Break

SESSION 8 11:00-12:45

SESSION 8: IN SITU TECHNIQUES  
ROOM: MAISTROS A  
CHAIR: A. CHATT, K. POTIRIADIS

11:00  
O8 A-1  
Itzhak Orion, Israel  
POTASSIUM TO NaCl RATIOS ASSESSMENTS IN THE DEAD SEA PONDS USING IN-SITU GAMMA SPECTROMETRY SYSTEM  
Itzhak Orion, Khalil Abu-Rabeah, Moti Aharoni, Oded Harel

11:15  
O8 A-2  
Effrosyni Androulakaki, Greece  
IN SITU gamma-RAY MEASUREMENTS ON THE SEABED IN TWO DIFFERENT MARINE ENVIRONMENTS  
Effrosyni Androulakaki, Christos Tsabaris, Georgios Eleftheriou, Michael Kokkoris, Dionissis Patiris, Filothei Pappa, Rosa Vlastou

11:30  
O8 A-3  
Dawn Wellman, USA  
ADVANCED SUBSURFACE 3D ELECTRICAL IMAGING AN 4D TIME-LAPSE MONITORING FOR GROUNDWATER CONTAMINANT REMEDIATION  
T. Johnson, Dawn Wellman

11:45  
O8 A-4  
Arturo Vargas, Spain  
RESPONSE STUDY OF A 1"x1" SPECTROMETRIC DOSE RATE LaBr3 MONITOR FOR GROUND DEPOSITED ARTIFICIAL RADIONUCLIDES  
Anna Camp, Arturo Vargas

12:00  
O8 A-5  
Lenka Thinova, Czech Republic  
LONG TERM MONITORING OF NPP TEMELIN (CR) OUTCOME RADIONUCLIDES USING LABORATORY AND IN SITU GAMMA SPECTROMETRY METHOD  
Lenka Thinova, Jaroslav Kluson, Kamila Johnova, Tomas Cechak

12:15  
O8 A-6  
Elder Magalhaes Souza, Brazil  
IN-SITU GAMMA RAY MEASUREMENTS FOR BACKGROUND ENVIRONMENTAL STUDIES AND CALCULATION OF EXTERNAL DOSES RATES IN BRAZIL  
Elder Magalhaes Souza, Elaine R.R. Rochedo, Claudio C. Conti

12:30  
O8 A-7  
Jacques Bezuidenhout, South Africa  
THE ANALYSES OF RADIATION RESULTS FROM VARIOUS GEOGRAPHICAL SETTINGS BY EMPLOYING A SELF-DEVELOPED IN SITU GAMMA RAY DETECTION SYSTEM  
Jacques Bezuidenhout

12:45 -13:30  
PL6  
PLENARY SESSION 6  
ROOM: MAISTROS A, CHAIR: A. IOANNIDOU & P.P. POVINEC

12:45  
PL6-1  
Françoise Brechignac, France  
RADIOECOLOGY FOR THE 21ST CENTURY – NEW TRENDS  
Françoise Brechignac

13:15  
Closing Ceremony



POSTER Presentations

Posters

SESSION 1 RECENT DEVELOPMENTS IN ANALYTICAL PS1 TECHNOLOGIES NUCLEAR ACCIDENTS ATMOSPHERIC RADIOACTIVITY

PS1-1 Erika Brattich, Italy EIGHT YEARS OF <sup>210</sup>Pb CONCENTRATIONS MONITORING AT TWO SITES IN THE MEDITERRANEAN REGION: SEASONALITY AND IDENTIFICATION OF POTENTIAL SOURCES Erika Brattich, Miguel Angel Hernández-Ceballos, Giorgia Cinelli, Juan Pedro Bolivar, Laura Tositti

PS1-2 Toshihiro Yoshihara, Japan MEASUREMENT OF ENVIRONMENTAL RADIOACTIVITY USING A CUMULATIVE GAMMA RADIATION DOSIMETER; FERTILIZATION INDUCED CHANGES IN YOUNG FRUIT TREES Toshihiro Yoshihara, Yuto Nagao, Shin-Nosuke Hashida, Naoki Kawachi, Nobuo Suzui, Yong-Gen Yin, Shu Fujimak

PS1-3 Alessandra Pacini, Brazil SIGNATURE OF A SUDDEN STRATOSPHERIC WARMING IN THE NEAR-GROUND <sup>7</sup>Be FLUX. Alessandra Pacini, Ilya Usoskin, Kalevi Mursula, Ezequiel Echer, Heitor Evangelista

PS1-4 Zarina Serzhanova, Kazakhstan SPECIATION OF TRITIUM IN SOIL AT THE TERRITORY OF THE SEMIPALATINSK TEST SITE Zarina Serzhanova, Almira Aidarkhanova, Sergey Lukashenko, Almira Raimkanova

PS1-5 Dominik Höweling, Germany BACKGROUND REDUCTION IN A GAMMA SPECTROMETER SETUP WITH AN ACTIVE SHIELDING Dominik Höweling, Bernd Hettwig and Helmut W. Fischer

PS1-6 Elisa Gordo, Spain DETECTION OF FUKUSHIMA-RELEASED RADIOISOTOPES IN MÁLAGA (SOUTH SPAIN) Elisa Gordo, Concepción Dueñas, Esperanza Liger, Sergio Cañete, Manuel Pérez, Carmen Fernández

PS1-7 Yasuto Miyake, Japan DESIGN OF THE NEGATIVE ION COOLER AT MALT Yasuto Miyake, Hiroyuki Matsuzaki, Shuichi Hasegawa

PS1-8 Claudia Grossi, Spain ANALYSIS OF THE RELATIONSHIP BETWEEN CLIMATIC PHENOMENA & ATMOSPHERIC VARIABILITY OF <sup>7</sup>Be AND <sup>210</sup>Pb CONCENTRATIONS IN NORTHEASTERN SPAIN Claudia Grossi, Isabel Serrano, Antonia Camacho, J. Morgui, X. Rodó, Maria Amor Duch

PS1-9 Esperanza Liger, Spain SURFACE ACTIVITY LEVELS OF <sup>7</sup>Be, <sup>210</sup>Pb, <sup>40</sup>K AND OTHER ATMOSPHERIC SPECIES AND THE INFLUENCE OF AFRICAN AIR MASS INTRUSIONS Esperanza Liger, Elisa Gordo, Concepción Dueñas, Sergio Cañete, Pedro Cañada, María Cabello Manuel Pérez

PS1-10 Hiroyuki Matsuzaki, Japan STUDY OF IODINE DYNAMICS IN SOIL BASED ON THE ANALYSIS AND MODELLING FOR THE I-129 DEPTH PROFILES BEFORE AND AFTER THE FDNPP ACCIDENT Hiroyuki Matsuzaki, Maki Honda, Yasuto Miyake, Marina Kawamoto, Hironori Tokuyama and Wataru Kijima

PS1-11 Suk Hyun Kim, Korea CONCENTRATIONS OF RADIOCESIUM FOR SEAFOOD IN KOREA, 2011-2014 Suk Hyun Kim, Gi Hoon Hong, Hyun Mee Lee, Hee Young Park, Young Sub Sim

PS1-12 Alyona Yankauskas, Kazakhstan NFLUENCE OF RADIONUCLIDES <sup>3</sup>H ON MORPHO-ANATOMY INDEX OF PLANTS ACHNATHERUM SPLENDENS AND ELYMUS ANGUSTUS ON THE STS Alyona Yankauskas, Natalya Larionova, Sergey Lukashenko, Fuad Nasyrov

PS1-13 Natalia Alegria - Spain BENCHMARKING OF MONTE CARLO SIMULATIONS FOR A LANTHANUM BROMIDE (LaBr3) DETECTOR Fernando Legarda and Natalia Alegria

PS1-14 Eleftheria Ioannidou, Greece THE INFLUENCE OF TROPOPAUSE HEIGHT IN <sup>7</sup>Be ATMOSPHERIC CONCENTRATIONS Ari-Pekka Leppanen, Eleftheria Ioannidou, Dimitrios Melas, Alexandra Ioannidou

PS1-15 Miguel Angel Hernandez Ceballos, Italy <sup>7</sup>Be ACTIVITY CONCENTRATIONS ON SURFACE AEROSOLS IN SPAIN – IDENTIFICATION AND METEOROLOGICAL ANALYSIS OF PEAK PERIOD Rafael Luis Lozano, Miguel Angel Hernandez Ceballos, Jose Antonio Adame, Giorgia Cinelli, Erika Brattich

PS1-16 Arturo Vargas, Spain AMBIENT DOSE EQUIVALENT RATE RESPONSE OF GAMMA SPECTROMETRY MONITORS IN THE FRAMEWORK OF METROERM EUROPEAN PROJECT Arturo Vargas, Anna Camp, Maria Roig, Maria Amor Duch, Merce Ginjaume, Ulrich Stöhlker

PS1-17 Sandra Regina Damatto, Brazil <sup>7</sup>Be CONCENTRATION IN AIR SURFACE OVER A LONG PERIOD OF MONITORING IN THE CITY OF SÃO PAULO, BRAZIL Sandra Regina Damatto, Marcelo Francis Máduar, Brigitte Roxana Pecequilo, Paulo Rene Nogueira, Marcelo Nisti

PS1-18 Pavel P. Povinec, Slovakia TRITIUM AND RADIOCARBON IN SEAWATER OFFSHORE FUKUSHIMA Pavel P. Povinec, Laval Liong Wee Kwong, Róbert Janovics, Jakub Katzer, Mikály Molnár, Masanao Nakano, László Palcsu, Mai K. Pham, Jean-Baptiste Philippe

PS1-19 Junwei Jia, USA BEEHIVE MODEL CREATION FOR USE IN DETERMINING RADIATION DOSE TO BEES AND BEE LARVAE Junwei Jia, Kathryn Higley, Emily Caffrey

PS1-20 Tetsuya Matsunaka, Japan STUDY ON THE MIGRATION BEHAVIOUR OF FUKUSHIMA ACCIDENT-DERIVED IODINE-I-129 FROM LAND AREA TO THE MARINE ENVIRONMENT Tetsuya Matsunaka, Kimikzau Sasa, Keisuke Sueki, Yuichi Onda, Takashi Ishimaru, Keisuke Taniguchi, Yoshifumi Wakiyama, Maki Honda, Tsutomu Takahashi, Masumi Matsumura, Hiroyuki Matsuzaki

PS1-21 Carmen Varlam, Romania TRITIUM LEVEL IN THE ENVIRONMENT AT CERNAVODA NUCLEAR POWER PLANT AFTER 15 YEARS OF OPERATION Carmen Varlam, Ionut Faurescu, Irina Vagner, Gheorghe Titescu, Elena Bobric, Ion Popescu, Vasile Simionov, Denisa Faurescu

PS1-22 Gheorghe Titescu, Romania TRITIUM LEVEL EVOLUTION IN THE ENVIRONMENT AT ICIT AFTER 5 YEARS OF MONITORING PROGRAMME Irina Vagner, Carmen Varlam, Ionut Faurescu, Denisa Faurescu, Gheorghe Titescu, Diana Bogdan

PS1-23 Alexandr Novikov, Kazakhstan INVESTIGATING RADIOLOGICAL SITUATION FEATURES AT P-1 TECHNICAL SITE Alexandr Novikov, Murat Umarov, Sergey Lukashenko, Yulia Yakovenko

PS1-24 Helen Papaefthymiou, Greece SPATIAL AND VERTICAL DISTRIBUTION OF <sup>137</sup>Cs IN A GREEK AREA FOLLOWING THE CHERNOBYL NUCLEAR REACTOR ACCIDENT Dimitra Dimitrellou, Helen Papaefthymiou, Eleni Zagana

PS1-25 Yuuta Terasaka, Japan ESTIMATION OF AIR AND SURFACE RADIOACTIVITY CONCENTRATIONS FROM PULSE HEIGHT DISTRIBUTION MEASURED BY NaI(Tl) SCINTILLATION DETECTOR Yuuta Terasaka, Jun Hirouchi, Shigekazu Hirao, Jun Morizumi, Hiromi Yamazawa and Yuu Kuwahara

PS1-26 Shigekazu Hirao, Japan ATMOSPHERIC DISPERSION AND SURFACE DEPOSITION OF RADIONUCLIDES DISCHARGED BY FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT ACCIDENT Shigekazu Hirao, Hiromi Yamazawa and Jun Morizumi

PS1-27 Ivan Sykora, Slovakia LONG-TERM VARIATIONS OF ATMOSPHERIC RADIONUCLIDES IN BRATISLAVA AIR Ivan Sykora, Pavel P. Povinec, Karol Holý and Miroslav Jeskovsky

PS1-28 Ivo Svetlik, Czech Republic THE VALLEY SYSTEM OF THE JIHLAVA RIVER AND MOHELNO RESERVOIR WITH ENHANCED TRITIUM ACTIVITIES Ivo Svetlik, Pavel Simek, Tereza Korínková, Pavel Povinec, Michal Fejgl, Irena Malátová, Lenka Tomaskova

PS1-29 Sang Han Lee, Korea DEVELOPMENT OF RICE REFERENCE MATERIAL AND A PROFICIENCY TEST FOR THE MEASUREMENT OF Cs-137 AND K-40 Sang Han Lee, Jung Suk Oh, Jong Man Lee, K. B Lee, Min Kie Lee, Yong Ho Choi

<b>PS1-30</b>	Miroslav Jeskovsky, Slovakia
<b>STUDY OF AEROSOL <sup>137</sup>Cs ACTIVITIES AROUND NUCLEAR POWER PLANTS IN SLOVAKIA</b>	
<i>Miroslav Jeskovsky, Pavel Povinec, Ondrej Slávik and Ivan Šýkora</i>	
<b>PS1-31</b>	Peter Szanto, Hungary
<b>APPLICATION OF THE ATMOSPHERIC DISPERSION MODELLING IN THE INTERMEDIATE AND LATE PHASE OF NUCLEAR ACCIDENT</b>	
<i>Peter Szanto, Lilla Hoffmann, Emese Homolya, Tamas Pazmandi, Marton Zagyvai and Peter Zagyvai</i>	
<b>PS1-32</b>	Dobromir Pressyanov, Bulgaria
<b>LABORATORY INFRASTRUCTURE FOR RADON AND THORON MEASUREMENTS BY CDS/DVDS</b>	
<i>Dobromir Pressyanov, Krasimir Mitev, Ivelina Dimitrova, Strahil Georgiev and Jordan Kolev</i>	
<b>PS1-33</b>	Robert Breier, Slovakia
<b>MONTE CARLO SIMULATION OF BACKGROUND OF HPGe DETECTORS IN DEEP UNDERGROUND INSTALLATIONS</b>	
<i>Robert Breier, Pavel Povinec and Wolfango Plastino</i>	
<b>PS1-34</b>	Denis Turchenko, Kazakhstan
<b>STUDY OF AIR BASIN NATURE AND RADIOACTIVE CONTAMINATION LEVELS AT THE SEMIPALATINSK TEST SITE (STS) AND THE ADJACENT TERRITORIES</b>	
<i>Denis Turchenko, Sergey Lukashenko and Assan Aidarkhanov</i>	
<b>PS1-35</b>	Jung Suk Oh, Korea
<b>SEQUENTIAL SEPARATION OF Pu ISOTOPES AND <sup>90</sup>Sr IN SEAFOOD USING ION-EXCHANGE/EXTRACTION CHROMATOGRAPHY</b>	
<i>Jung Suk Oh, Sang Han Lee, Jong Ki Choi, Sung Hwan Kim, Hyung Soo Kim, Ji-Yeon Kwak, Sun Hee Park</i>	
<b>PS1-36</b>	Sofija Forkapic, Serbia
<b>FIRST RADIOACTIVITY MONITORING OF SOIL IN SERBIA</b>	
<i>Dragana Todorovic, Sofija Forkapić, Milica Rajacic, Kristina Bikit, Natasa Sarap and Dusan Mrdja</i>	
<b>PS1-37</b>	Maria Angelica Wasserman, Brazil
<b>MULTI-CRITERIA OPTIMIZATION TOOL FOR THE DECONTAMINATION OF URBAN AREAS AFTER A NUCLEAR ACCIDENT</b>	
<i>Elaine R.R. Rochedo, Diogo Neves Silva, Christiano Luca, Maria Angelica Wasserman, Pedro R.R. Rochedo, Jean Remy Guimarães, Ricardo Tadeu Lopes</i>	
<b>PS1-38</b>	Alena S. Petrova, Russia
<b>EFFECT OF TRITIUM ON BIO-AND PHOTOLUMINESCENCE OF BLUE FLUORESCENT PROTEIN</b>	
<i>Alena S. Petrova, Anna A. Lukonina, Nadezhda S. Kudryasheva</i>	
<b>PS1-39</b>	Kimikazu Sasa, Japan
<b>ESTIMATION OF I-131 DEPOSITION FROM I-129 ANALYSIS IN SURFACE SOILS RELEASED FROM THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT</b>	
<i>Kimikazu Sasa, Masumi Matsumura, Tetsuya Matsunaka, Tsutomu Takahashi, Yukihiko Satou, Norikazu Kinoshita, Hiroyuki Matsuzaki, Keisuke Sueki</i>	
<b>PS1-40</b>	Jaroslav Kluson, Czech Republic
<b>AIRBORNE MONITORING IN THE COURSE OF NUCLEAR ACCIDENT SCENARIOS AND IN POST ACCIDENTAL ENVIRONMENTAL CONDITIONS</b>	
<i>Jaroslav Kluson, Tomáš Urban</i>	
<b>PS1-41</b>	Sarata Kumar Sahoo, Japan
<b>Sr-90 MEASUREMENT IN JAPANESE SOIL SAMPLES AFTER THE FUKUSHIMA NUCLEAR ACCIDENT</b>	
<i>Sarata Kumar Sahoo, Norbert Kavasi, Jerzy Wojciech Mietelski, Atsuyuki Sorimachi, Edyta Lokas, Tatsuo Aono</i>	
<b>PS1-42</b>	Esperanza Liger, Spain
<b>ATMOSPHERIC DEPOSITION CHARACTERISTICS OF <sup>7</sup>Be AT MÁLAGA (SOUTH SPAIN)</b>	
<i>Esperanza Liger, Concepción Dueñas, Elisa Gordo, Sergio Cañete, María Cabello, Manuel Pérez</i>	
<b>PS1-43</b>	Miroslav Hýža, Czech Republic
<b>REMOTE CONTROLLED AEROSOL SAMPLER WITH GAMMA SPECTROMETRIC MODULE – DESIGN, FUNCTIONALITY AND INITIAL WORKING EXPERIENCE</b>	
<i>Miroslav Hýža, Petr Rulík, Martin Fiala, Lukáš Skála</i>	
<b>PS1-44</b>	Jussi Paatero, Finland
<b><sup>210</sup>Pb IN SIZE-SEGREGATED AEROSOL SAMPLES IN NORTHERN FINLAND</b>	
<i>Jussi Paatero, Alexandra Ioannidou and Jukka Lehto</i>	

## SESSION 2

## PS2

## PROGRAM IN ENVIRONMENTAL RADIOACTIVITY STUDIES NATURAL RAADIONUCLIDES IN THE ENVIRONMENT

<b>PS2-1</b>	Jovana Nikolov, Serbia
<b>DETERMINATION OF PANNONIAN BASIN (SERBIA) GROUNDWATER AGES USING TRITIUM RADIOACTIVE ISOTOPE</b>	
<i>Jovana Nikolov, Tanja Petrović Pantić, Nataša Todorovic, Milan Tomic, Ines Krajcar Bronic, Jadranka Barešić, Kristina Bikit, Ivana Stojković</i>	
<b>PS2-2</b>	Dimitris Mitrakos, Greece
<b>RADON CONCENTRATION IN A NATURALLY VENTILATED RADIOACTIVE WASTE INTERIM STORAGE FACILITY IN GREECE</b>	
<i>Dimitris Mitrakos, Maria Kolovou, Maria Nikolaki, Anastasia Savidou and Constantinos Potiriadis</i>	
<b>PS2-3</b>	Alexandra Stepanenko, Russia
<b>β/γ-TRACER OF DANGEROUS NATURAL PHENOMENA AND TECHNOGENIC ACCIDENTS</b>	
<i>Alexandra Stepanenko, Valentina Yakovleva, Natalia Mishina, Petr Nagorskiy, Xenia Ryabkina</i>	
<b>PS2-4</b>	Jing Chen, Canada
<b>AN ESTIMATE OF CANADIAN POPULATION EXPOSURE TO NATURAL BACKGROUND TERRESTRIAL RADIATION</b>	
<i>Jing Chen, Ken Ford, John Buckle and John Carson</i>	
<b>PS2-5</b>	Mario Enrique Gomez Fernandez, USA
<b>CREATION AND APPLICATION OF VOXELIZED DOSIMETRIC MODELS: AN EVALUATION OF ABSORBED FRACTIONS IN APIS MELLIFERA</b>	
<i>Mario Enrique Gomez Fernandez, Kathryn Higley</i>	
<b>PS2-6</b>	Constantin Cosma, Romania
<b>RADON STUDIES IN FIVE SHOW KARSTIC CAVES FROM ROMANIA</b>	
<i>Constantin Cosma, Denissa Bety Burghеле, Botond Papp, Alexandra Cucos, Silviu Constantin, Alexandru Petculescu and Oana Moldovan</i>	
<b>PS2-7</b>	Erinda Ndrecka, Albania
<b>ANALYTICAL INVESTIGATION OF POTTERY FROM DIFFERENT NEOLITHIC PERIOD IN SOUTH-EAST OF ALBANIA</b>	
<i>Erinda Ndrecka, Nikolla Civici, Ilir Gjipali</i>	
<b>PS2-8</b>	Fotini Noli, Greece
<b>DISTRIBUTION AND SEASONAL VARIATION OF RADIOACTIVITY, MINOR AND TRACER ELEMENTS IN LAKES IN THE LIGNITE MINING AREA OF NORTH-WESTERN GREECE</b>	
<i>Fotini Noli, Panagiotis Tsamos</i>	
<b>PS2-9</b>	Claudia Grossi, Spain
<b>APPLICATION OF THE DILUTION COEFFICIENT OF <sup>222</sup>Rn FOR AIR QUALITY STUDIES OF PM10 IN A NORTH-WESTERN SPANISH CITY</b>	
<i>Claudia Grossi, Manuel Dall’Osto, Miguel Angel Hernandez Ceballo, Marco Pandolfi, Arturo Vargas</i>	
<b>PS2-10</b>	Margarita Herranz, Spain
<b>COMPARISON OF SEVERAL METHODS FOR THORIUM-ISOTOPES DETERMINATION IN ENVIRONMENTAL AND INDUSTRIAL SAMPLES</b>	
<i>Margarita Herranz, Juan Carlos Lozano, Juan Pedro Bolivar, Rafael Garcia Tenorio</i>	
<b>PS2-11</b>	Saroa Rozas, Spain
<b>PUBLIC EXPOSURE TO ENVIRONMENTAL RADIOACTIVITY IN BILBAO (SPAIN)</b>	
<i>Saroa Rozas, Raquel Idoeta, Margarita Herranz, Fernando Legarda</i>	
<b>PS2-12</b>	Nataliya Mishina, Russia
<b>SEASONAL VARIATIONS OF RADIATION TRACERS IN THE GROUND ATMOSPHERE</b>	
<i>Nataliya Mishina, Valentina Yakovleva, Petr Nagorskiy, Alexandra Stepanenko, Kseniya Ryabkina, Maxim Cherepnev</i>	
<b>PS2-13</b>	Juan Mantero, Sweden
<b>USE OF REFERENCE MATERIALS RGU1 AND RGTh1 IN ENVIRONMENTAL GAMMA SPECTROMETRY MEASUREMENTS</b>	
<i>Juan Mantero, Santiago Hurtado and Rafael Garcia-Tenorio</i>	
<b>PS2-14</b>	Fernando Mosqueda, Spain
<b>RADIOACTIVE CHARACTERIZATION OF LEACHATES AND EFFLORESCENCES IN A PHOSPHOGYPSUM DISPOSAL SITE AS A PRELIMINARY STEP BEFORE ITS RESTORATION</b>	
<i>Fernando Mosqueda, Manuel Jesús Gázquez González, Silvia Pérez Moreno, Rafael Garcia-Tenorio, Juan Pedro Bolivar, Juan Mantero Cabrera, Elisa Rodríguez López</i>	
<b>PS2-15</b>	Viviana Mossa, Italy
<b>IONIZATION CHAMBER OF NEW CONCEPTION FOR ENVIRONMENTAL GAS RADON MEASUREMENTS</b>	
<i>Viviana Mossa, Giuseppe Roselli, Claudia Monte, Francesco Barile, Luigi Vitucci, Vincenzo Paticchio, Luigi Schiavulli</i>	
<b>PS2-16</b>	Nadri Mohamed, Algeria
<b>VERTICAL PROFILE OF 137CS, 210 PB AND 40K IN GHARDIA REGIONS, ALGERIA</b>	
<i>Nadri Mohamed, Ioannidou Alexandra and Khiari Chams-Eddine</i>	

<b>PS2-17</b>	Juan Carlos Lozano, Spain
<b>AN ARRANGEMENT FOR THE STUDY OF DISTRIBUTION COEFFICIENTS IN SOILS NATURALLY ENRICHED IN URANIUM ISOTOPES AND <sup>226</sup>Ra</b>	
<i>Juan Carlos Lozano, Adriana De Souza Medeiros Batista, Cristina Prieto Calvo, Pilar Blanco Rodríguez, Feliciano Vera Tomé</i>	
<b>PS2-18</b>	Nerantzis Kazakis,, Greece
<b>THE URANIUM ISOTOPE IN THE CHARACTERIZATION OF GROUNDWATER IN ANTHEMOUNTAS RIVER BASIN, NORTHERN GREECE</b>	
<i>Nerantzis Kazakis, George Vargemезis, Alexandra Ioannidou and Fotini Noli</i>	
<b>PS2-19</b>	Xeniya Ryabkina, Russia
<b>INFLUENCE OF PRECIPITATION ON FLUXES OF BETA AND GAMMA RADIATION IN THE GROUND ATMOSPHERE</b>	
<i>Xeniya Ryabkina, Pyotr Nagorskiy, Valentina Yakovleva, Maxim Cherepnev, Alexandra Stepanenko, Nataliya Mishina</i>	
<b>PS2-20</b>	Evangelos Giagias, Romania
<b>RADON INDOOR CONCENTRATION AND ITS SEASONAL VARIATION IN ATHENS, GREECE</b>	
<i>Evangelos Giagias, Constantin Cosma, Delia Marcu, Denisa Burghel</i>	
<b>PS2-21</b>	Nevenka Antovic, Montenegro
<b>POTASSIUM-40 IN THE COASTAL AREA OF MONTENEGRO – DOSE RATE ASSESSMENT</b>	
<i>Nevenka Antovic, Nikola Svrkota, Ivanka Antovic, Ranko Svrkota, Gordana Lastovicka-Medin</i>	
<b>PS2-22</b>	Jing Chen, Canada
<b>A STUDY ON THE CORRELATION BETWEEN SOIL RADON POTENTIAL AND AVERAGE INDOOR RADON POTENTIAL IN CANADIAN CITIES</b>	
<i>Jing Chen, Ken Ford</i>	
<b>PS2-23</b>	Rana Baydoun, Lebannon
<b>MEASUREMENT OF NATURAL AND ARTIFICIAL RADIOACTIVITY IN THE LEBANSE ENVIRONMENT FOR TOTAL DOSE ASSESSMENT</b>	
<i>Omar El Samad, Rana Baydoun, Wissam Zaidan, Hamzeh El Jeaид, Rola Alayan</i>	
<b>PS2-24</b>	Arturo Vargas, Spain
<b>OUTDOOR RADON MONITORING NETWORK TO STUDY THE BEHAVIOUR OF <sup>222</sup>Rn OVER SPAIN</b>	
<i>Claudia Grossi, Arturo Vargas, Anna Camp, José Enrique Martín, Juan Pedro Bolivar Josep, Anton Morgui</i>	
<b>PS2-25</b>	Joao Arruda-Neto, Brazil
<b>EVALUATION OF URANIUM IN ORGANS OF RESIDENTS FROM AN URANIUM-RICH REGION USING TEETH AS BIOINDICATORS</b>	
<i>Joao Arruda-Neto, Henriette Righi, Yadirа Medіna, Marcos Antonio Cascino, Fermin Garcia, Godofredo Genofre</i>	
<b>PS2-26</b>	Ioanna Liatsou, Cyprus
<b>RADIUM CONCENTRATION IN (TE)NORM MATERIALS BY RADON EMANATION PRIOR AND AFTER ACIDIC DISSOLUTION OF THE SAMPLES</b>	
<i>Ioanna Liatsou, Ioannis Pashalidis</i>	
<b>PS2-27</b>	Young Gun Ko, Korea
<b>DEVELOPMENT AND VALIDATION OF A METHODOLOGY FOR DETERMINATION OF U AND Th IN SOIL USING ALPHA SPECTROMETRY</b>	
<i>Young Gun Ko, Jong-Myoung Lim, Hoon Lee, Young-Ji, Kun Ho Chung, Mun Ja Kang, Geun-Sik Choi</i>	
<b>PS2-28</b>	Young-Yong Ji, Korea
<b>METHOD VALIDATION FOR THE QUANTIFICATION OF NATURAL RADIONUCLIDES USING A GAMMA-RAY SPECTROMETRY IN RAW MATERIALS AND BY-PRODUCTS</b>	
<i>Young-Yong Ji, Chang-Jong Kim, Kun Ho Chung, Jong-Myooung Lim, Young Gun Ko, Hyuncheol Kim Mun Ja Kang</i>	
<b>PS2-29</b>	Paulo Silva, Brazil
<b>EFFECTIVE DOSE ASSESSMENT DUE TO ÁGUAS DE LINDÓIA WATER INGESTION</b>	
<i>Paulo Silva, Catia Saueia</i>	
<b>PS2-30</b>	Ioanna Sfampa, Greece
<b>INTEGRATED DOSES FROM NATURAL RADIOACTIVITY MEASUREMENTS BY USING LIF:Mg,Cu,P AND AL2O3:C THERMOLUMINESCENCE DOSIMETERS (TLD)</b>	
<i>Ioanna Sfampa, Eleni Theodoridou, Stylianos Stoulos and George Kitis</i>	
<b>PS2-31</b>	Irene Lopes, Portugal
<b>RADON (Rn-222) DETERMINATION IN PORTUGUESE WATERS USING TWO PHASE LIQUID SCINTILLATION COUNTING</b>	
<i>Irene Lopes, João Abrantes, Albertina Libânio, Maria José Madruga and Mário Reis</i>	
<b>PS2-32</b>	Cucos Alexandra, Romania
<b>INDOOR RADON EXPOSURE IN SOME ENERGY- EFFICIENT HOUSES FROM ROMANIA</b>	
<i>Cucos Alexandra, Cosma Constantin and Evangelos Giagias</i>	

<b>PS2-33</b>	Maria Sotiropoulou, Greece
<b>APPLICATION OF THE ERICA ASSESSMENT TOOL FOR THE CALCULATION OF THE DOSE RATES IN TERRESTRIAL HERBIVORE MAMMALS IN GREECE</b>	
<i>Maria Sotiropoulou, Heleny Florou, Metaxia Manolopoulou</i>	
<b>PS2-34</b>	M.F.Maduar, Brazil
<b>EXTERNAL AND INTERNAL ANNUAL EFFECTIVE DOSES FOR PARANÁ STATE GRANITES USED AS INTERNAL COATING BUILDING MATERIALS BY HIGH RESOLUTION GAMMA-RAY SPECTROMETRY</b>	
<i>A.O. Ferreira, B.R.S.Pecequilo, M.F.Maduar</i>	
<b>PS2-35</b>	Konstantinos Hatzzioannou, Greece
<b>ABSORBED DOSE ESTIMATION TO FAMILY MEMBERS OF PATIENTS TREATED WITH RADIOIODINE</b>	
<i>Konstantinos Hatzzioannou, Kosmas Badiavas, Emmanouil Papanastasiou, Panagiota Koutla, Marios Velonis and Ioannis Iakovou</i>	
<b>PS2-36</b>	Ilektra Kotopoulou, Greece
<b>NATURAL RADIOACTIVITY LEVELS IN THE SHALLOW-SEA HYDROTHERMAL SYSTEM OFF SE MILOS ISLAND, GREECE</b>	
<i>Ilektra Kotopoulou, Theo J. Mertzimekis, Athanasios Godelitsas, Roy Price, David A. Fike, Jan P. Amend, William P. Gilhooly Iii and Greg K. Druschel</i>	
<b>PS2-37</b>	Sang Han Lee, Korea
<b>DEVELOPMENT OF RICE REFERENCE MATERIAL AND A PROFICIENCY TEST FOR THE MEASUREMENT OF Cs-137 AND K-40</b>	
<i>Sang Han Lee, Jung Suk Oh, Jong Man Lee, K. B Lee, Min Kie Lee, Yong Ho Choi</i>	
<b>PS2-38</b>	Jongmyoung Lim, Korea
<b>METHOD VALIDATION FOR THE QUANTIFICATION OF RADIOACTIVITY CONCENTRATION IN NORM SAMPLES USING ICP-MS</b>	
<i>Jongmyoung Lim</i>	
<b>PS2-39</b>	Sheldon Landsberger, USA
<b>ON USING NON-DESTRUCTIVE NEUTRON ACTIVATION ANALYSIS TO EFFECTIVELY DETERMINE <sup>235,238</sup>U, <sup>40</sup>K AND <sup>232</sup>Th IN VARIOUS MATRICES WITH SUB-GRAM QUANTITIES OF MATERIAL</b>	
<i>Sheldon Landsberger, Richard Lara</i>	
<b>PS2-40</b>	Theodoros Mertzimekis, Greece
<b><sup>137</sup>Cs RETAINMENT IN CENTRAL GREECE SOILS: A STUDY UNDERTAKEN A RADIOCESIUM HALFLIFE AFTER CHERNOBYL</b>	
<i>Theodoros Mertzimekis, Ioannis Ioannidis, Athanasios Godelitsas, Dionisis Gasparatos, Konstantinos Stamoulis and Konstantinos Ioannides</i>	
<b>PS2-41</b>	Elaine R.R. Rochedo, Brazil
<b>ASSESSMENT OF THE EXTERNAL EXPOSURE OF THE BRAZILIAN POPULATION TO NATURAL BACKGROUND RADIATION</b>	
<i>Elaine R.R. Rochedo, Krause Salles, Elder de Souza, Pedro R.R. Rochedo and Maria Angelica Wasserman</i>	
<b>PS2-42</b>	Hiroki Sugiura, Japan
<b>OBSERVATION AND MODELLING OF ATMOSPHERIC RADON-222 TRANSPORT IN EAST ASIA</b>	
<i>Hiroki Sugiura, Shigekazu Hirao, Jun Moriizumi, Hiromi Yamazawa</i>	
<b>PS2-43</b>	Marcelo Francis Máduar, Brazil
<b>RADIATION HAZARD INDICES IN THE APPLICATION OF PHOSPHOGYPSUM MIXTURES AS A BUILDING MATERIAL: PROPOSAL FOR A REGULATION</b>	
<i>Marcelo Francis Máduar, Barbara P. Mazzilli, Marcelo B. Nisti</i>	

<b>SESSION 3</b>	<b>MODELING OF ENVIRONMENTAL PROCESSES</b>
<b>PS3</b>	<b>RADIONUCLIDE TRANSPORT – MARINE RADIOACTIVITY</b>
<b>PS3-1</b>	Nassim Bachir-Bey, Belgium
<b>ORIGIN OF HALIDES (Cl- AND Br-) AND OF THEIR STABLE ISOTOPES (d37Cl AND d81Br) AT THE TOURNEMIRE CLAYROCK URL (FRANCE) - EXPERIMENTAL AND NUMERICAL APPROACH</b>	
<i>Nassim Bachir-Bey</i>	
<b>PS3-2</b>	Santiago Miguel Enamorado Báez, Spain
<b>EFFECT OF PHOSPHOGYPSUM AMENDMENTS IN THE U AND Th UPTAKE AND PARTITIONING IN COTTON CROPS GROWN IN RECLAIMED SOILS FROM THE GUADALQUIVIR MARSHLANDS (SW SPAIN)</b>	
<i>Santiago Enamorado Báez, Jose María Abril Hernandez, Antonio Delgado García Andreu Cáceres</i>	
<b>PS3-3</b>	Kil Yong Lee, Korea
<b>GROUNDWATER RECHARGE OF INJECTION-PUMPING SYSTEM FOR WATER-CURTAIN HEATING SYSTEM USING RADON MASS BALANCE</b>	
<i>Kil Yong Lee, Yongchul Kim and Soo Young Cho</i>	
<b>PS3-4</b>	Natalya Larionova, Kazakhstan
<b>BIOLOGICAL SITES FOR MONITORING OF RADIATION HAZARDOUS OBJECTS</b>	
<i>Natalya Larionova, Sergey Lukashenko, Oksana Lyakhova and Almira Aidarkhanova</i>	

**PS3-5** Lucio Leonardo, Brazil  
**PRELIMINARY RESULTS OF <sup>238</sup>U, <sup>232</sup>Th AND TRACE ELEMENTS DETERMINED IN SOIL AND SEDIMENT PROFILES COLLECTED IN PONTE NOVA RESERVOIR, SÃO PAULO, BRAZIL**  
*Sandra Damatto, Lucio Leonardo, Marcelo Máduar, André Silva, Joseilton Souza,Barbara Mazzilli*

**PS3-6** Ivan Sanchez-Castro, Spain  
**SCREENING OF BACTERIAL STRAINS ISOLATED FROM URANIUM MILL TAILINGS POREWATERS FOR BIOREMEDIATION PURPOSES**  
*Ivan Sanchez-Castro, Ahinara Amador-Garcia, Cristina Moreno-Romero, Margarita Lopez-Fernandez, Fadwa Jroundi, Vannapha Phrommavanh, Michael Descostes and Mohamed Larbi Merroun*

**PS3-7** Juan Pedro Bolivar, Spain  
**HYDROGEOCHEMISTRY AND NATURAL RADIOACTIVITY OF A COMPLEX KARST SYSTEM: CASE STUDY OF SIERRA DE GÁDOR (ALMERÍA, SE SPAIN)**  
*José Luis Guerrero Márquez, Juan Pedro Bolivar, Fernando Mosqueda, Silvia Pérez Moreno, Ángela Vallejos, Francisco Sánchez Martos*

**PS3-8** L. Currivan, Ireland  
**IMPACT OF RADIOACTIVE WASTE DISCHARGES TO THE IRISH SEA: A STUDY OF ARTIFICIAL RADIOACTIVITY CONCENTRATIONS IN CARLINGFORD LOUGH, IRELAND**  
*L. Currivan, A. Semenov, L. Leon-Vintro*

**PS3-9** Jing Chen, Canada  
**A STUDY ON THE LEVELS OF RADIOACTIVITY IN FISH SAMPLES FROM EXPERIMENTAL LAKES AREA IN CANADA**  
*Jing Chen, Baki Sadi, Weihua Zhang, Nadereh Sr-Amant, Michael Rennie*

**PS3-10** Natasa Todorovic, Serbia  
**OPTIMIZATION OF LOW-LEVEL LIQUID SCINTILLATION COUNTER FOR <sup>90</sup>Sr DETERMINATION IN WATER SAMPLES USING CERENKOV RADIATION**  
*Natasa Todorovic, Ivana Stojkovic, Jovana Nikolov, Miodrag Krmar*

**PS3-11** Ivana Stojkovic, Serbia  
**COLOR QUENCH CORRECTION FOR GROSS ALPHA/BETA MEASUREMENTS IN WATERS**  
*Ivana Stojković, Nataša Todorovic,Jovana Nikolov*

**PS3-12** Maria Evangelia Souti, Germany  
**MEDICAL <sup>131</sup>I IN RIVER WATER AND SEDIMENTS CASE STUDY OF WESER RIVER IN NW GERMANY**  
*Maria Evangelia Souti and Helmut Fischer*

**PS3-13** Almira Raimkanova, Kazakhstan  
**RESEARCH OF LEACHING OF ARTIFICIAL RADIONUCLIDES FROM SOIL OF THE «BALAPAN» TESTING SITE**  
*Almira Raimkanova, Almira Aidarkhanova, Sergey Lukashenko and Zarina Serzhanova*

**PS3-14** Ivanka Antovic, Serbia  
**FIRST DETERMINATION OF Be-7 IN FISH FROM THE SOUTH ADRIATIC: LIZA SPECIES (L. aurata, L. ramada, L. saliens)**  
*Ivanka Antovic, Nikola Svrkota, Mirzeta Hadzibrahimovic, Nevenka Antovic, Ranka Zizic*

**PS3-15** Ferdinand Sudbrock, Germany  
**THE EFFECTIVENESS OF WASTEWATER TREATMENT IN NUCLEAR MEDICINE - PERFORMANCE DATA AND RADIOECOLOGICAL CONSIDERATIONS**  
*Ferdinand Sudbrock, Klaus Schomaecker, Alexander Drzezza*

**PS3-16** Loïc Ducros, France  
**ORGANICALLY BOUND TRITIUM ACTIVITIES WITHIN FRENCH CONTINENTAL WATERSHEDS - PRELIMINARY RESULTS FROM AREAS MOST IMPACTED BY PAST NUCLEAR FALLOUT**  
*Loïc Ducros, Frédérique Eyrolle-Boyer, Sabine Charmasson, Hervé Thébault, David Claval*

**PS3-17** Sang Han Lee, Korea  
**A STUDY OF ARTIFICIAL RADIONUCLIDES IN LAKE SEDIMENTS**  
*Sung Hwan Kim, Sang Han Lee, Jung Suk Oh, Jong Ki Choi and Taegu*

**PS3-18** Lucio Leonardo, Brazil  
**ACTIVITY CONCENTRATIONS OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN FELINE DRY FOOD**  
*Fernanda Cavalcante and Brigitte Roxana Soreanu Pecequilo, Lucio Leonardo*

**PS3-19** José Lourenço Friedmann Angeli,, Brazil  
**RADIOANALYTICAL ASSESSMENT OF SEDIMENTATION RATES IN THE CARAVELAS ESTUARY (BAHIA, BRAZIL) USING UNSUPPORTED <sup>210</sup>Pb AND <sup>137</sup>Cs MODELING**  
*José Friedmann Angeli, Natalia Venturini, Paulo Alves Lima Ferreira, Rubens César Lopes Figueira*

**PS3-20** Wagner de Souza Pereira, Brazil  
**URANIUM IN SURFACE WATER AT A NATURALLY OCCURRING RADIOACTIVE MATERIAL AREA IN BRAZIL**  
*Wagner de Souza Pereira, Alphonse Kelecom and Ademir Xavier Da Silva*

**PS3-21** Zuhaf Er, Turkey  
**TOTAL ALPHA-BETA RADIOACTIVITY OF BOTTLED DRINKING WATER IN TURKEY**  
*Zuhaf Er*

**PS3-22** Tereza Ježková, Czech Republic  
**DETERMINATION OF THE RADIONUCLIDE SOIL-TO-PLANT TRANSFER FACTORS - LABO RATORY METHODOLOGY**  
*Tereza Ježková, Petr Rulík*

**PS3-23** Manuel Perez Mayo, Germany  
**DATING OF A GERMAN RIVERINE LAKE SEDIMENT USING Pb-210, Cs-137 AND Be-7**  
*Manuel Perez Mayo, Daniela Pittauer, Helmut W. Fischer*

**PS3-24** Ivanka Lovrencic Mikelic, Croatia  
**VARIATION OF SEDIMENTATION RATE IN THE SEMI-ENCLOSED BAY DETERMINED BY Cs-137 DISTRIBUTION IN SEDIMENT (KAŠTELA BAY, CROATIA)**  
*Ivanka Lovrencic Mikelic*

**PS3-25** Murat BelivermiŞ Turkey  
**BIOACCUMULATION OF RADIOCESIUM IN ISOCHRYSIS GALBANA AND PHAEODACTYLUM TRICORNUTUM AND ITS ASSIMILATION IN MANILA CLAM RUDITAPES PHILIPPINARUM**  
*Narin Sezer, Murat Belivermis, Önder Kılıç, Gülsah Kalaycı, Kubilay Dogan Kılıç*

**PS3-26** Naglaa Fahmi, Egypt  
**ESTABLISHING A BASE LINE FOR RADIOACTIVITY IN ALEXANDRIA, EGYPT**  
*Naglaa Fahmi, Ahmed Khedr, Ahmed Mandour*

**PS3-27** Hedvig Simon, Romania  
**FLOODING EVIDENCE FOR THE LAST 30 YEARS IN DIFFERENT TYPES OF LAKES FROM ROMANIA**  
*Hedvig Simon, Robert-Csaba Begy, Szabolcs Kelemen*

**PS3-28** Alexandr Moshkov, Kazakhstan  
**RESEARCH OF ARTIFICIAL RADIONUCLIDES DISTRIBUTION IN PLACES WHERE ULTRA LOW YIELD NUCLEAR EXPLOSION AND NON-NUCLEAR EXPLOSIVE EXPERIMENTS WERE CARRIED OUT**  
*Alexandr Moshkov, Sergey Lukashenko, Murat Umarov, Alexandr Novikov*

**PS3-29** Galina Lujaniene, Lithuania  
**APPLICATION OF GRAPHENE OXIDE AND MAGNETIC GRAPHENE OXIDE FOR REMOVAL OF RADIONUCLIDES AND HEAVY METALS FROM CONTAMINATED WASTEWATER**  
*Galina Lujaniene, Sergej Šemcuk*

**PS3-30** R. García-Tenorio, Spain  
**RADIOACTIVE CHARACTERIZATION OF PRODUCED WATERS FROM GAS INDUSTRY IN MEXICO**  
*C.D. Mandujano-García, J. Mantero, A. Benavides-Mendoza, F. Martell-Valles, M. Sosa, G. Manjon, R. García-Tenorio*

**PS3-31** Vigilija Cidzikiene, Lithuania  
**TRACER EXPERIMENTS IN GROUNDWATER AT NEW NUCLEAR POWER PLANT SITE IN LITHUANIA**  
*Vigilija Cidzikiene, Vaidote Jakimaviciute Maseliene*

**PS3-32** Masatoshi Yamada, Japan  
**TEMPORAL CHANGE OF Pu INVENTORY IN WATER COLUMN OF THE BERING SEA**  
*Masatoshi Yamada and Jian Zheng Masatoshi Yamada, Jian Zheng*

**PS3-33** Raul Villegas, Brazil  
**NORM ASSESSMENT IN WATER TREATMENT SYSTEMS - POÇOS DE CALDAS/BR CASE**  
*Raul Villegas, Henrique Fukuma, Rafael de Moura, Adriano Ferrerira*

**PS3-34** Chiara Cantaluppi, Italy  
**THREE YEARS MONITORING OF GAMMA-EMITTING RADIONUCLIDES IN FISHING PRODUCTS SOLD IN ITALIAN MARKET AND ARISING FROM MAIN FAO FISHING AREAS**  
*Chiara Cantaluppi, Federica Ceccotto, Andrea Fasson*

PS3-35	Yihong Xu, China
PLUTONIUM CHARACTERISTICS IN SEDIMENTS OF ROMO COASTAL AREA IN DENMARK	
Yihong Xu, Xiaolin Hou, Jixin Qiao, Shaoming Pan	
PS3-36	Irina Vagner, Romania
METHOD FOR ORGANICALLY BOUND TRITIUM ANALYSIS FROM SEDIMENT USING A COMBUSTION BOMB	
Irina Vagner, Carmen Varlam, Ionut Faurescu, Denisa Faurescu, Diana Bogdan	
PS3-37	Dawn Wellman, USA
RISK-INFORMED PRIORITIZATION AND SYSTEMS-BASED APPROACHES FOR REMEDIATION OF DEEP SUBSURFACE CONTAMINATION	
Dawn Wellman, M. Truex, M. Freshley, H Lee, J Morse, Skip Chamberlain, Kurt Gerdes	
PS3-38	Flavia Groppi, Italy
AMAD OF <sup>7</sup> Be AEROSOL UNDER DIFFERENT METEOROLOGICAL CONDITIONS AND DIFFERENT ENVIRONMENTS	
Flavia Groppi, Simone Manenti, Luigi Gini, Alexandra Ioannidou	
PS3-39	Jordi Fons, Spain
SIMULTANEOUS DETERMINATION OF SPECIFIC ALPHA AND BETA EMITTERS BY LSC-PLS IN WATER SAMPLES	
Jordi Fons, Montse Llauradó	
PS3-40	Philipp Steinmann, Switzerland
DETECTION OF Ra-223 FROM THERAPEUTIC APPLICATIONS IN MUNICIPAL SEWAGE SLUDGE	
Philipp Steinmann	
PS3-41	Istvan Bikit, Serbia
CORRELATIONS BETWEEN SOIL CHARACTERISTICS AND RADIOACTIVITY CONTENT OF VOJVODINA SOIL	
Sofija Forkapic, Jovica Vasin, Istvan Bikit, Dusan Mrdja, Kristina Bikit and Stanko Milic	
PS3-42	Pilar Blanco-Rodriguez, Spain
INFLUENCE OF SOIL TEXTURE AND AVAILABILITY ON THE SOIL-TO-PLANT TRANSFER OF <sup>238</sup> U AND <sup>226</sup> Ra	
Pilar Blanco-Rodriguez, Feliciano Vera-Tomé, Juan Carlos Lozano	
PS3-43	Almira Aidarkhanova, Kazakhstan
INVESTIGATION OF DISTRIBUTION OF RADIOACTIVE CONTAMINATION IN THE "WATER – SEDIMENTS" SYSTEM OF THE SEMIPALATINSK TEST SITE AND ADJACENT TERRITORIES	
Almira Aidarkhanova, Sergey Lukashenko	
PS3-44	Flavia Groppi, Italy
FALLOUT MEASUREMENTS IN NORTH ITALIAN REGIONS AFTER FUKUSHIMA NUCLEAR POWER PLANT ACCIDENT	
Flavia Groppi, Simone Manenti, Luigi Gini, Alexandra Ioannidou	



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# PROGRAM AT A GLANCE

	Monday 21 September	Tuesday 22 September	Wednesday 23 September	Thursday 24 September	Friday, 25 September
08:30	Registration	Poster Session 1	Poster Session 2		Poster Session 3
09:00	Opening	Plenary Session 2 PL2	Plenary Session 3 PL3	Plenary Session 4 PL4	Plenary Session 5
09:30	Plenary Session 1 PL1				
10:00		Coffee Break	Coffee Break	Coffee Break	Coffee Break
10:30					
11:00		Coffee break			
11:30			Session 5A:GROUND WATER Session 5B:RADIOECOLOGY I	Session 6A:ATMOSPHERE Session 6B:RADIOECOLOGY II	Session 8 IN SITU TECHNIQUES
12:00		Session 1A:AMS Session 1B:MARINE			Plenary Session 6
12:30			Excursion		Closing Ceremony
13:00	Lunch	Lunch		Lunch	
13:30					
14:00	Poster Session 1	Poster Session 2		Poster Session 3	
14:30					
15:00	Session 2A: RADIOANALYTICS Session 2B: AQUATIC	Session 4A: SOIL Session 4B: NORM/Radon		Session 7A:ATMOSPHERE Session 7B:RADIOECOLOGY III	
15:30					
16:00					
16:30	Coffee break	Coffee break		Coffee break	
17:00	Session 2A: RADIOANALYTICS Session 2B: AQUATIC	Session 4A: SOIL Session 4B: NORM/Radon		Session 7A: ATMOSPHERE Session 7B: RADIOECOLOGY IIII	
17:30					
18:00	Honorary Session				
18:30					

# New Frontiers for Accelerator Mass Spectrometry - New Isotopes and New Applications

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Keywords: Actinides, <sup>236</sup>U, AMS, <sup>135</sup>Cs, <sup>93</sup>Zr  
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Despite numerous applications, AMS is usually restricted to a handful of isotopes (<sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, <sup>36</sup>Cl, <sup>41</sup>Ca, <sup>129</sup>I). Our goal at the Vienna Environmental Research Accelerator (VERA) is to develop detection methods for more isotopes and to extend the scope of the method by new applications.

AMS is the mass spectrometric method with the highest abundance sensitivity, which is a prerequisite for the measurement of the long-lived radioisotope <sup>236</sup>U ( $t_{1/2}$ =23.4 million years). Over the last years, the Vienna Environmental Research Accelerator (VERA) was continuously extended to optimize its detection.

The most successful application so far is oceanography, since anthropogenic <sup>236</sup>U is present in the world oceans at <sup>236</sup>U:<sup>238</sup>U levels from 10<sup>-11</sup> to 10<sup>-8</sup>. We have explored methods to increase the sensitivity and thus to reduce the water volume required to 1 L or less, which significantly reduces the sampling effort. This increased sensitivity is also necessary to address the expected typical natural isotopic ratios on the order. Other actinides (Pu, Np, Am) profit from this development.

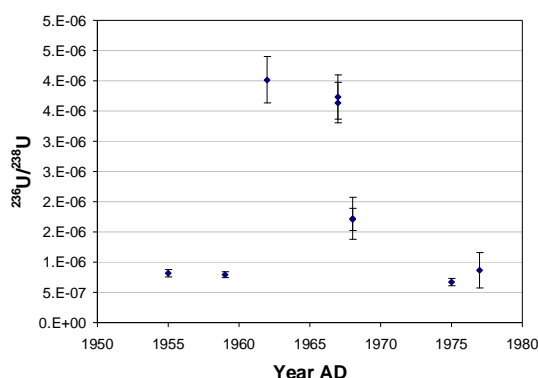


Figure 1 <sup>236</sup>U/U ratios were as high as 10<sup>-6</sup> in the bomb test era.

Samples originate from freshwater, ocean water, corals, deep sea sediments, soil, peat, air filters, and the biosphere. The fields of applications are mainly environmental tracing, nuclear forensics, and radiation protection. We will present results on <sup>236</sup>U from the Fukushima exclusion zone, from a sediment-buried peat bog, and from roe deer antlers from the bomb peak era (Figure 1). First results on materials expected to be unaffected by anthropogenic <sup>236</sup>U suggest that this contamination is more widespread than expected, and

that improved chemical procedures have to be developed to fully exploit the instrumental limit.

The optimized setup of VERA is robust against chemical impurities in the background, which e.g. allows measuring Pu isotopes directly in a uranium matrix. This can significantly simplify chemical sample preparation for actinide detection, which because actinides can be separated altogether from the matrix and measured in the same AMS sample. This method was verified using waters from several sources.

Fission products are widely used for tracer studies of environmental processes, in nuclear forensics, and are important for nuclear waste disposal. Presently, <sup>129</sup>I is the only fission product measured with AMS. Besides the well-known <sup>137</sup>Cs ( $T_{1/2}$ =30yr), the longer-lived sister isotope <sup>135</sup>Cs ( $T_{1/2}$ =2.3Myr) is of special interest, because the combination of the two isotopes would allow for identifying sources of contamination. Because of its long half-life, <sup>135</sup>Cs cannot be detected via decay counting. The stable isobar <sup>135</sup>Ba presently prevents AMS measurements. Similarly, <sup>93</sup>Zr with a half-life of 1.5×10<sup>6</sup> a is also produced with high fission, but suffers from the interference of the stable isobar <sup>93</sup>Nb. We hope to achieve isobar suppression by Laser photodetachment, the process of neutralizing a negative ion with a laser beam. By careful selection of the photon energy, only unwanted isobars are neutralized and selectively suppressed. We present a preparatory study on the performance of the VERA AMS facility for <sup>135</sup>Cs and <sup>93</sup>Zr. To our surprise, the discrimination of <sup>93</sup>Zr performed significantly better than expected from simulations. Assuming realistic numbers for chemical Niobium reduction, a detection level of <sup>93</sup>Zr/Zr below 10<sup>-9</sup> seems feasible even without photodetachment.

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## Applications of $^{14}\text{C}$ and $^{129}\text{I}$ to some problems in environmental science

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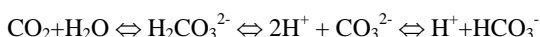
Keywords: carbon-14, iodine-129, Fukushima, lakes.

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There are many useful environmental applications of the radionuclides  $^{14}\text{C}$  and  $^{129}\text{I}$ , which can be easily measured by accelerator mass spectrometry. These two nuclides have very different sources:  $^{14}\text{C}$  ( $t_{1/2}$  5,730 yr) is mainly the result of secondary nuclear reactions in the upper atmosphere due to cosmic-ray effects, although there is a recent component of  $^{14}\text{C}$  due to nuclear testing (Hua et al. 2013). By contrast,  $^{129}\text{I}$  is mainly a fission product ( $t_{1/2}$  15.7 Myr), and is an excellent tracer of recent anthropogenic sources.

Carbon-14 in freshwater: The study of  $^{14}\text{C}$  in freshwater systems, particularly lakes is complex due to the many pathways that can exist in a freshwater ecosystem. We define dissolved inorganic carbon (DIC), meaning mainly carbonates and bicarbonates in solution in fresh waters. These species are generally in equilibrium, such that:



Where the left-hand species are more abundant at lower pH and the bicarbonate species predominate at the highest pH. This equilibrium works in artificial solutions, however it is not entirely correct in natural waters. At pH>8, that calculated values of  $\text{CO}_2$  greatly exceed measured values. This is also true at low pH, for soft waters and bogs. This difference is in part due to interferences by organic acids and other organic constituents which disturb the purely inorganic equilibrium, by slowing down the creation of carbonate and also the dissociation of  $\text{HCO}_3^-$  to  $\text{CO}_2$ .

Dissolved organic carbon defines a wide range of species, which are operationally defined as passing a <1µm filter. They include fulvic acids, which are water-soluble acids at any pH and humic acids, which are higher molecular weight than fulvic acids and are base-soluble but acid-insoluble (Thurman, 1985). Other DOC components might include low-molecular-weight acids, carbohydrates, lipids, aromatics, plant and animal particulates. There are many different ways to extract DOC from water systems, a summary is given in table 2 (Leonard et al. 2013): vacuum drying and subsequent

combustion of the organic residues, solid-phase extraction, UV oxidation and ultrafiltration.

We will present some results on a study of  $^{14}\text{C}$  in two very disparate environments: a complex system at Lake Qinghai in China and local surface water in Tucson, Arizona.

Iodine-129: At the Arizona AMS Laboratory, we have developed a number of projects focusing on  $^{129}\text{I}$ . These studies focus on both ocean studies, particularly related to the Fukushima nuclear accident, and also to the development of  $^{129}\text{I}$  as a possible tracer of modern groundwater recharge. The Fukushima study includes monitoring of  $^{129}\text{I}$  in Pacific Ocean water samples collected on a regular basis since 2011 at the Scripps Institution of Oceanography in La Jolla, CA, other coastal locations in California and also sample collection in Kaoshiung, Taiwan. This monitoring has been undertaken since shortly after the Fukushima event in 2011. Our results show the arrival of an enhanced  $^{129}\text{I}$  signal in mid-2014. Our newer study is concentrated on water in semi-arid environments. We have begun a small pilot project to demonstrate the possible usefulness of  $^{129}\text{I}$  as an age-tracer of recent recharge to shallow aquifers in the Tucson Basin. Iodine-129 concentrations have been measured in precipitation, surface water, and groundwater samples. Initial results from this study and also the on-going Pacific Ocean  $^{129}\text{I}$  study related to the Fukushima event will be presented.

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# Isotopes and Climate Change

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## Introduction

Climate change is one of the most discussed subjects in our time, both in science and in the public. As is well known, these discussions range from the scientific-based IPCC reports claiming that human-induced climate changes are already happening to flat-out denials that anthropogenic impact is relevant for climate changes.

We do know that during the history of the Earth climate has changed by natural processes in sometimes dramatic ways (e.g. the coming and going of ice ages). It is probably reasonable to assume that the climate was not influenced by man significantly until the last 200 years or so, when the industrial revolution and the population growth literally changed the face of the Earth. This period is therefore coined the *Anthropocene* by Crutzen and Stoermer (2000), signifying man's influence on the Earth system in many domains. While some changes are obvious, climate change in general depends on a complex interplay between different natural variables, and now we have man's contribution as well.

The concept of isotopes (Soddy, 1913) and the first separation of stable neon isotopes by mass spectrometry (Thomson 1913) presented us for the last 100 years with a unique tool to study the environment at large by reading and interpreting the "isotope language". Here we will discuss a few examples of how this language can be utilized for climate change studies.

## Stable Isotopes

The power of stable isotopes to study climate change comes from mass fractionation, i.e. the dependence of physical, chemical and biological processes on the mass of isotopes, changing thereby the isotopic abundances within an element. Particularly useful are isotope ratios of light elements, e.g.  $^2\text{H}/^1\text{H}$ ,  $^{13}\text{C}/^{12}\text{C}$ ,  $^{18}\text{O}/^{16}\text{O}$  and  $^{15}\text{N}/^{14}\text{N}$  usually expressed in the  $\delta$  notation. For  $\delta^{18}\text{O}$ , this denotes the relative deviation of the  $^{18}\text{O}/^{16}\text{O}$  ratio from some internationally fixed standard value and is expressed in units of permil (‰). The  $\delta$  values are widely used as proxies to study a variety of important parameters (e.g. temperature) in the environment.

For example, the first  $\delta^{18}\text{O}$  record in a Greenland ice core gave a proxy for the temperature during the last 100,000 years (Dansgaard et al., 1969). Later this was extended back to 740,000 years in Antarctica (EPICA et al., 2004), supporting the astronomical forcing theory of Milankovic which had also been observed in the marine  $\delta^{18}\text{O}$  record (Bassinot et al., 1994).

As another example, the interesting question whether the appearance and disappearance of the Norse in Greenland (1000-1400 AD) was linked to

climatic changes has been investigated with  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in bones, reflecting possibly climate-driven changes in dietary food. (Nelson et al., 2011-2012).

## Long-lived Radioisotopes

Accelerator mass spectrometry (AMS) provides a method to study climate changes with long-lived cosmogenic and anthropogenic radioisotopes (Kutschera, 2010). The capability to measure radioisotope-to-stable isotope ratios from  $10^{-12}$  to  $10^{-16}$ , allows one to trace climate changes with unprecedented sensitivity. In addition to mass fractionation, time information can be gained from radioisotope measurements.

By far the most-used radioisotope in AMS is  $^{14}\text{C}$ . A big issue in climate change is  $\text{CO}_2$ , considered to be the most important anthropogenic contribution to global warming (Broecker 1975). With AMS tens of thousands of  $^{14}\text{CO}_2$  measurements in the world oceans have been performed [McNichol et al., 2000], which helped to refine global ocean carbon climatology (Key et al., 2004).

The second most-used radioisotope in AMS is  $^{10}\text{Be}$ , which is produced by cosmic-ray secondaries in surfaces of rocks. Its accumulation with time is being used extensively for surface exposure dating, tracing the waxing and waning of glaciers throughout the Holocene (e.g. Ivy-Ochs et al., 2009).

## Conclusion

Only a few selected example of the use of isotopes are mentioned above. AMS with its enormous breadth of applications (Kutschera, 2013), is particularly well suited for investigations in connection with climate change. In the current presentation, a comprehensive overview of the power of the isotope language to study climate change will be given.

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# Comparison of $^{129}\text{I}/^{127}\text{I}$ ratio in fish and seawater samples from the Pacific Ocean

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Keywords:  $^{129}\text{I}/^{127}\text{I}$  ratio, tracer, fish, seawater, ocean biology.

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The proportion of  $^{129}\text{I}$  (HL: 15.7 Myr) and  $^{127}\text{I}$  (stable) ( $^{129}\text{I}/^{127}\text{I}$ ) was the order of  $10^{-12}$  in the pre-nuclear era (Hou et al 2009). But in the nuclear era, much amount of anthropogenic  $^{129}\text{I}$  as well as other radioactive nuclides have been released into the Earth's surface environment. The  $^{129}\text{I}/^{127}\text{I}$  ratio in recent surface seawater has risen up to  $10^{-6}$  (He et al, 2013). The highest  $^{129}\text{I}/^{127}\text{I}$  ratio was observed in the European North Sea resulted by direct discharge of  $^{129}\text{I}$  from nuclear reprocessing plants. On the other hand, in the Pacific Ocean, far from the currently active source,  $^{129}\text{I}$  is transported through the atmosphere, resulting the  $^{129}\text{I}/^{127}\text{I}$  ratio of  $10^{-11}$ - $10^{-9}$ . The  $^{129}\text{I}/^{127}\text{I}$  ratio has also a strong depth-dependency. Since anthropogenic  $^{129}\text{I}$  diffused from the surface to the deeper layer and  $^{129}\text{I}/^{127}\text{I}$  ratio showed decreasing depth profile (e.g. Povinec et al, 2010). These variations mean that  $^{129}\text{I}/^{127}\text{I}$  ratio characterizes each ocean location.

Iodine is known to be a biophile element. For example, iodine is essential element for fish's thyroid metabolism (Jarque and Piña, 2014). The  $^{129}\text{I}/^{127}\text{I}$  ratio of ocean biota would depend on that of seawater in the habitat environment. Therefore the  $^{129}\text{I}/^{127}\text{I}$  ratio has potential to be a tracer of ocean biology. In this work the  $^{129}\text{I}/^{127}\text{I}$  ratio in fish and seawater samples were measured and compared to verify the relationship of  $^{129}\text{I}/^{127}\text{I}$  ratio between fishes and seawater.

Fish and seawater samples were collected at the Sta. NBD (47°N, 160°E) and Sta. SBD (25°N, 160°E) in the Pacific Ocean by the research cruise KH-14-2, 2014. The 2 benthic and 4 bathypelagic fish samples were collected by the beam trawl (ca. 5,200 m depth) at the Sta. NBD. The 2 bathypelagic fish samples were collected by the plankton net (170 m depth) at the Sta. SBD.

Detailed treatment procedure is shown in Kusuno et al (2015). Iodine in fish samples was extracted by pyrohydrolysis and then it was purified by solvent extraction. Iodine in seawater samples was purified by solvent extraction. The Woodward iodine (Woodward iodine co.) carrier was added to both fish and seawater samples. Measurements of  $^{129}\text{I}/^{127}\text{I}$  ratio were carried out with accelerator mass spectrometry at the Micro Analysis Laboratory, Tandem Accelerator, the University of Tokyo. The  $^{127}\text{I}$  concentration in sample was measured by ICP-MS (Agilent 7500a) to calculate  $^{129}\text{I}/^{127}\text{I}$  ratio of the samples.

Fig. 1 shows the  $^{129}\text{I}/^{127}\text{I}$  ratio of the fish and seawater samples from the Sta. NBD and the Sta. SBD. The  $^{129}\text{I}/^{127}\text{I}$  ratio of fish samples was ranged  $(1.5\text{--}14.8) \times 10^{-11}$ . These  $^{129}\text{I}/^{127}\text{I}$  ratio was consistent with that in

surface seawater (ca.  $3 \times 10^{-11}$ ) regardless of whether fishes are bathypelagic or benthic. This suggests 1)  $^{129}\text{I}/^{127}\text{I}$  ratio of fishes was not controlled directly by seawater, and 2)  $^{129}\text{I}/^{127}\text{I}$  ratio in fishes are controlled by something surface components. This may have relevance with i)  $\text{I}^-$  is concentrated in surface seawater (Ito et al 2003) and ii)  $\text{IO}_3^-$  isn't directly taken by biota (Shaw 1962).

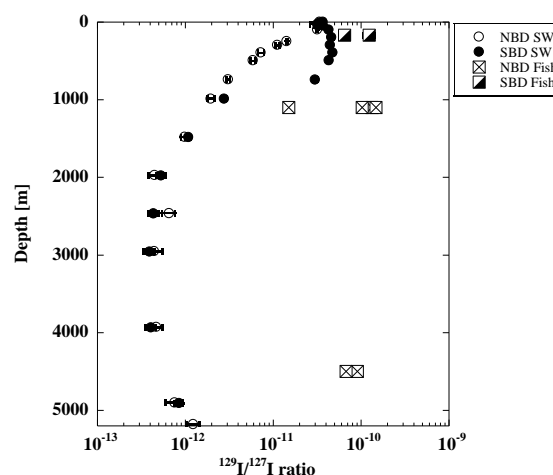


Figure 1. The  $^{129}\text{I}/^{127}\text{I}$  ratio of fish and seawater samples from the Pacific Ocean.

## Acknowledgments

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# Iodine-129 in coral cores from the Philippines as a tracer of anthropogenic nuclear activities

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**Keywords:** I-129, coral, ocean-atmosphere circulation, radionuclide transport, environment

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Iodine-129 is the longest-living radioisotope of iodine having a half-life of 15.7 Ma. While produced naturally through cosmic ray spallation of xenon in the atmosphere and spontaneous fission of uranium in the earth's crust, this signal has since been dwarfed by <sup>129</sup>I from human nuclear activities such as nuclear weapons testing, nuclear fuel reprocessing, and nuclear accidents.

While the massive release of radionuclides in the environment is unfortunate, <sup>129</sup>I effectively became an attractive option for tracer applications, particularly in the marine environment, due to its long half-life, long oceanic residence times (400,000 years), and because its release points are generally known and in the case of nuclear reprocessing plants, continuous.

This study investigates <sup>129</sup>I/<sup>127</sup>I in coral cores – one of the most prominent records of physical/chemical properties in seawater. This is to explore its application as a historical tracer of anthropogenic nuclear activities and atmosphere-ocean circulation and transport.



Figure 1. Philippine current (Baler, Aurora) and future (Spratly Islands) study sites.

Specifically, coral cores were obtained from the Philippines (Fig. 1), an interesting study site since its eastern side directly receives signal from the Pacific Ocean (where US nuclear weapons test sites are located), while its western side is isolated to a certain extent. Moreover, this is one of the first historical records of <sup>129</sup>I from corals in the northern hemisphere, where majority of nuclear activities occur.

<sup>129</sup>I was measured using Accelerator Mass Spectrometry (AMS) and <sup>127</sup>I (stable iodine) by Inductively-Coupled Plasma Mass Spectrometry (ICP-MS).

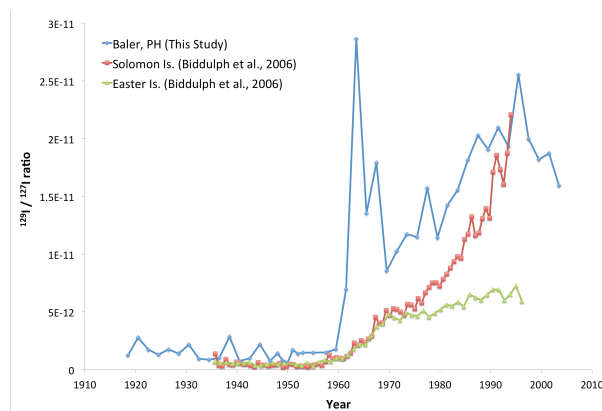


Figure 2. <sup>129</sup>I/<sup>127</sup>I ratio in corals from the Philippines (Baler, PH) and the southern hemisphere corals – Solomon and Easter Islands (Biddulph et al., 2006).

Results show more enhanced <sup>129</sup>I signal in corals from the Philippines compared to corals from the southern hemisphere (Fig. 2). A sharp <sup>129</sup>I bomb peak at around 1960s is observed, largely attributed to US nuclear weapons testing in the Pacific. Timing of the signal suggests a 5-year transit time from the Pacific weapons testing site to the Philippines. Post-bomb peak profile mainly reflects European nuclear fuel reprocessing activities but also possibly the Chernobyl nuclear accident (1995 peak) and additional weapons tests by China (region around 1975-1990). In contrast, these signals have an 11-year lag time, suggesting a different pathway – first through the atmosphere then a longer route around the Pacific Ocean. Furthermore, pre-bomb peaks show natural <sup>129</sup>I levels and variations. These appear to have similarities with coral oxygen-18 records, which generally reflect sea surface temperatures. Further studies differentiating signals are needed to provide more insights to these findings.

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# Changes in $^{129}\text{I}/^{127}\text{I}$ ratio of crater lake and volcanic activity at Zao volcano, Japan

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The volcanic activity has become higher at Zao volcano in Miyagi and Yamagata Prefectures, Japan, since January 2013 after the 2011 Tohoku Earthquake (JMA, 2015). Up to 106 times of the monthly volcanic earthquakes were observed in August 2014, and twice of the white turbidity were found in the surface of crater lake in October 2014. Basic water quality of crater lake have been studied by Tohoku University since the water quality of crater lake are correlating with volcanic activity. As a part of this investigation, we are trying to monitor the volcanic activity using  $^{129}\text{I}/^{127}\text{I}$  ratios at Zao volcano.

Long-lived  $^{129}\text{I}$  ( $T_{1/2} = 15.7$  million years) is produced in nature by cosmic-ray-induced spallation of xenon in the atmosphere and spontaneous fission of uranium in the lithosphere (Hou et al., 2009). Values of  $^{129}\text{I}/^{127}\text{I}$  ratios found nowadays in different environments are 2–8 orders of magnitude larger than that in the pre-nuclear era ( $1.5 \times 10^{-12}$ , Fehn et al., 2007) by the release of anthropogenic  $^{129}\text{I}$  originating from spent nuclear fuel reprocessing, nuclear weapons testing, and nuclear accident (Hou et al., 2009).  $^{129}\text{I}/^{127}\text{I}$  ratio in precipitation at Fukushima area rose to  $\sim 10^{-5}$  soon after the nuclear accident (Xu et al., 2013). In our previous study,  $^{129}\text{I}/^{127}\text{I}$  ratio in lake water collected in October 2013 from the crater lake at Zao volcano were  $2.2 \times 10^{-9}$ , which were affected by anthropogenic  $^{129}\text{I}$ . In terms of the global iodine cycle, chronologically-old iodine with low isotopic ratio was considered to be supplied into the crater lake from underground corresponding to the volcanic activity, resulting the decrease in  $^{129}\text{I}/^{127}\text{I}$  ratio of the crater lake. The present study aimed to elucidate distribution of  $^{129}\text{I}/^{127}\text{I}$  ratio and source of iodine in the crater lake, and the relativity between  $^{129}\text{I}/^{127}\text{I}$  ratio in the crater lake and volcanic earthquake for the monitoring of volcanic activity at Zao volcano using  $^{129}\text{I}/^{127}\text{I}$  ratio.

Water samples (2,000 ml) were collected 6 times from the surface of crater lake in eastern side of Zao volcano from October 2013 to October 2014 including before and after the white turbidity phenomena. In May 2014, water samples were also collected from the central lake in water depths of 0 m, 6 m, 12 m, and 18 m at the site of  $\sim 20$  m water depth, snow ice around the lake, and Zao hot spring. After adding 2 mg iodine carrier to the filtered water sample of 1,000 ml, the iodine was isolated and precipitated as AgI. The  $^{129}\text{I}/^{127}\text{I}$  ratio of AgI target was measured using an AMS system at the Micro Analysis Laboratory Tandem Accelerator (MALT), The University of Tokyo.  $^{127}\text{I}$  in the water sample was measured by an ICP-MS at the University of Tsukuba. The original  $^{129}\text{I}/^{127}\text{I}$  ratios in the water samples were calculated using  $^{127}\text{I}$  concentration obtained from ICP-MS and  $^{129}\text{I}/^{127}\text{I}$  ratio obtained from AMS.

$^{129}\text{I}/^{127}\text{I}$  ratios of the crater lake were  $(0.4\text{--}5.6) \times 10^{-9}$ , 2–3 orders of magnitude larger than that of Zao hot

spring  $((3.2\text{--}5.7) \times 10^{-12})$  which is higher contribution of iodine from the underground, and 1–2 orders of magnitude lower than that of snow ice  $((1.0\text{--}1.3) \times 10^{-8})$  including the anthropogenic  $^{129}\text{I}$ . Relatively high correlation between  $^{129}\text{I}/^{127}\text{I}$  ratios and  $1/^{127}\text{I}$  was found in the three types of water ( $R^2 = 0.69$ ,  $n = 13$ ), and the depth profile of  $^{129}\text{I}/^{127}\text{I}$  ratios in the crater lake was decrease with depth. Therefore, iodine in the crater lake consider to originate primary from the meteoric water and the underground, the  $^{129}\text{I}/^{127}\text{I}$  ratios in the lake were determined by the mixing of both sides.

$^{129}\text{I}/^{127}\text{I}$  ratios of the crater lake increased from  $2.2 \times 10^{-9}$  to  $5.6 \times 10^{-9}$  during October 2013 to the middle of October 2014, then, abruptly decrease to  $4.3 \times 10^{-10}$  soon after the second white turbidity (Fig. 1). A relatively strong negative correlation was found between the  $^{129}\text{I}/^{127}\text{I}$  ratio and monthly volcanic earthquake ( $R^2 = 0.68$ ,  $n = 4$ ), there is a possibility that the  $^{129}\text{I}/^{127}\text{I}$  ratio in the crater lake was related to the volcanic activity at Zao volcano. Further investigations are needed for the monitoring of the volcanic activity.

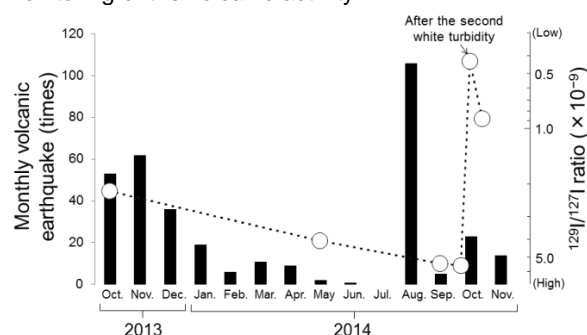


Figure 1 Temporal changes in  $^{129}\text{I}/^{127}\text{I}$  ratio of crater lake (white circles) and monthly volcanic earthquake (black bar, JMA, 2015) at Zao volcano.

## Acknowledgments

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# Long-lived cosmogenic radionuclides: Determination by accelerator mass spectrometry and model applications

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**Introduction:** Long-lived radionuclides with half-lives of 0.1-16 Ma (Tab. 1) have nowadays thousands of exciting applications, especially within environmental and geosciences. In nature, the so-called **cosmogenic nuclides (CNs)** are products of nuclear reactions induced by primary and secondary cosmic rays. Hence, they can be found in **extraterrestrial material** such as meteorites - originating from the asteroid belt, the Moon or Mars - and lunar samples in higher concentrations (e.g.  $\sim 10^{10}$  <sup>10</sup>Be atoms/g or  $< 0.5$  mBq/g). A combination of several CNs is used to reconstruct the exposure history of this unique material while in space (irradiation age) and on Earth (terrestrial age).

Though, in terrestrial material the concentrations are typically only on the order of  $10^4$ - $10^9$  atoms/g (i.e.  $\mu$ Bq/g - nBq/g) for <sup>10</sup>Be produced in the Earth's atmosphere, so-called **atmospheric or meteoric** <sup>10</sup>Be, transported to the surface and further absorbed and incorporated at and in, e.g. sediments or ice. Some of the lowest <sup>10</sup>Be concentrations ( $\sim 10^3$  atoms/g), produced **in-situ** by neutron- and muon-induced nuclear reactions from e.g. oxygen and silicon in quartz, can be found in samples taken from the Earth's surface. The concentrations of atmospheric or in-situ produced CNs record information to reconstruct sudden geomorphological events such as volcanic eruptions, rock avalanches, tsunamis, meteor impacts, earthquakes and glacier movements. Additionally, glacier movements and data from ice cores give hints for the reconstruction of historic climate changes and providing information for the validation of climate model predicting future changes. Slower processes such as sedimentation, river incision and erosion rates can also be investigated and last but not least, indirect dating of bones as old as several Ma's is possible.

**Anthropogenic** production by release from nuclear reprocessing, accidents and weapons testing led to increased levels of CNs in surface water and soil (<sup>129</sup>I,...), ice (<sup>36</sup>Cl,...) and of course, material from nuclear installations themselves (<sup>41</sup>Ca,...).

Some of the CNs can be further used as **natural or artificial tracers** to follow pathways in oceanography, to date and identify sources of groundwater, to perform retrospective dosimetry and to study aspects in radioecology, phytology, nutrition, toxicology and pharmacology.

**Method:** Today, the analytical method of choice for long-lived cosmogenic radionuclides – especially non- $\gamma$ -active ones - is **accelerator mass spectrometry (AMS)**. In contrast to decay counting, AMS scientists do not wait for the disintegration of the radioactive nucleus. In fact, the not-yet-decayed radionuclides are identified more

efficiently by mass spectrometry. The main advantage of using a high-energy accelerator for mass spectrometry is the nearly complete **elimination of background and interfering signals**, resulting from molecular ions and ions with similar masses e.g. isobars. Thus, AMS generally provides much **lower detection limits** in comparison to conventional mass spectrometry. Our DREAMS (DREsden AMS) system (Akhmadaliev et al., 2013) offers excellent measurement capabilities also for external users.

Table 1. Radionuclides measured by AMS at DREAMS.

Nuclide	$t_{1/2}$ [Ma]	Nuclide ratios of samples [ $10^{-12}$ ] (machine blank level)
<sup>10</sup> Be	1.387	0.01-300 ( $5 \times 10^{-16}$ ) <sup>10</sup> Be/ <sup>9</sup> Be
<sup>26</sup> Al	0.705	0.001-60 ( $8 \times 10^{-16}$ ) <sup>26</sup> Al/ <sup>27</sup> Al
<sup>36</sup> Cl	0.301	0.007-700 ( $2 \times 10^{-16}$ ) <sup>36</sup> Cl/ <sup>35</sup> Cl
<sup>41</sup> Ca	0.104	0.02-9000 ( $8 \times 10^{-15}$ ) <sup>41</sup> Ca/ <sup>40</sup> Ca
<sup>129</sup> I	15.7	artificial samples ( $3 \times 10^{-14}$ ) <sup>129</sup> I/ <sup>127</sup> I
actinides		under development

The benefits from using AMS are obvious and manifold: Smaller sample sizes, easier and faster sample preparation, higher sample throughput and the redundancy for radiochemistry laboratories are largely reducing costs. Lower detection limits widen applications to shorter and longer time-scales and to sample types that could never be investigated before. Nevertheless, **basic but accurate radiochemical sample separation** is an essential prerequisite for AMS measurements.

**Model applications:** Some of the first successful CN-projects performed at DREAMS had been:

- Dating of **marine sediments** (with ANU, ETH, TANDAR, TUM & VERA) by <sup>10</sup>Be & <sup>26</sup>Al and search for supernova-origin <sup>60</sup>Fe (by AMS at ANU & TUM)
- Growth rates of **deep-sea manganese nodules** by <sup>10</sup>Be and <sup>26</sup>Al (with Senckenberg)
- <sup>41</sup>Ca-determination in **water and concrete** from a **nuclear power plant** by LSC and AMS (with VKTA)
- Reconstruction of **meteorites'** history by <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, <sup>41</sup>Ca (with U Poznan & Bern, MPI Mainz,...).

## Acknowledgments

Thanks to all brave DREAMS-users working with a newly installed AMS-facility and for help from colleagues at other AMS-facilities (ANSTO, ANU, ASTER, ETH, VERA...) with cross-measurements and setting-up the time-of-flight-system for future actinide measurements.

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# Detection of Np and Pu in Sea Water with regard to the Fukushima Accident using AMS

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Keywords: AMS, actinides, extraction chromatography, ocean water

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Reports on the detection of plutonium (Pu) in litter samples collected in Fukushima Prefecture showed a  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio larger than 0.3 [1], which is in good agreement with reactor model calculations for the Fukushima Daiichi Nuclear Power Plant [2]. Consequently, a possible entry of Pu in the environment by the reactor accident can be distinguished from global fallout which shows a  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio of 0.18 [3]. If an emission into the Pacific Ocean water is considered, tropospheric close-in fallout from the Pacific Proving Grounds which can have isotopic ratios up to  $^{240}\text{Pu}/^{239}\text{Pu} = 0.34$  [3] has also to be taken into account. In order to identify Pu emitted into the Pacific Ocean by the reactor accident, the  $^{241}\text{Pu}/^{239}\text{Pu}$  ratio has to be determined in addition. Due to its short half-life ( $T_{1/2} = 14.325$  a),  $^{241}\text{Pu}$  originating from the bomb tests has already decayed by a large fraction.

For the study of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{237}\text{Np}$  concentrations in Pacific Ocean water samples taken at different locations and depths, the ultra-sensitive detection technique of accelerator mass spectrometry (AMS) was chosen. The detection of  $^{241}\text{Pu}$  by AMS is very challenging as its environmental concentration is estimated to be as low as  $0.5 \cdot 10^6$  atoms/kg(water) and because of isobaric interference from its daughter nucleus  $^{241}\text{Am}$ . Therefore, a highly effective chemical procedure for elemental separation for the AMS had to be implemented at the Maier-Leibnitz-Laboratory (MLL) in Munich. A procedure based on extraction chromatography using TEVA<sup>TM</sup> resin [4] was optimized for the application to the Pacific Ocean water samples. As the use of HF is off-limits in our laboratory, this reagent had to be eliminated from the separation method. For the estimation of the chemical yields, the procedure had to meet also the requirements of  $\alpha$  and  $\gamma$  spectrometry and ICP-MS, apart from the demands of AMS.

In a set of experiments at the Radiochemie München (RCM) the procedure was optimized such that the Pu and Np fractions were obtained with high chemical yield and Am was sufficiently suppressed at the same time. As tracers 20 Bq of  $^{243}\text{Am}$  and  $^{239}\text{Np}$  and 2 Bq of  $^{242}\text{Pu}$  solution, provided by the RCM, were used. The respective activities in the final fractions were determined by  $\alpha$  ( $^{243}\text{Am}$ ,  $^{242}\text{Pu}$ ) and  $\gamma$  spectrometry ( $^{239}\text{Np}$ ). In addition, Pu and Np yields were determined by ICP-MS at the Helmholtz-Zentrum Dresden-Rossendorf using  $10^{12}$  atoms of  $^{242}\text{Pu}$  and  $^{237}\text{Np}$  as spikes. The developed method was applied to analyse the concentration of Pu and Np in the certified reference material, IAEA-443, by AMS at the MLL to check the applicability of the method to sea water samples.

The experiments showed high chemical yields of around 90 % for Np and Pu and an Am suppression of  $10^4$ . In a first measurement campaign of the IAEA-443 samples with AMS it was not possible to analyse the concentration of  $^{239}\text{Pu}$  due to the strong interference of  $^{238}\text{U}$ . For an additional U suppression in the Pu fraction, the procedure was extended by a UTEVA column. The corresponding results of the AMS measurements are presented in Fig.1 which are in good agreement with the literature values [5].

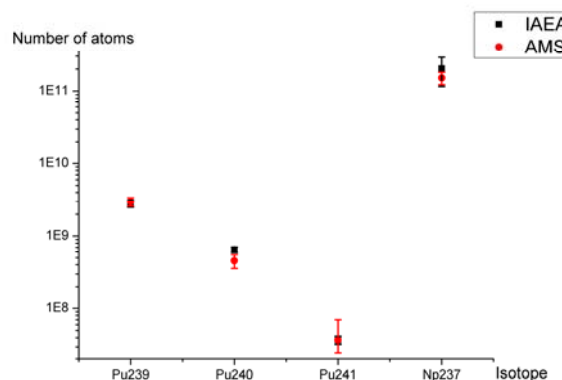


Figure 1: Literature values (black squares) and values detected by AMS (red dots) for the amount of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{237}\text{Np}$  in IAEA-443 Atlantic water with 1 sigma uncertainties.

The modified procedure is currently being applied to a first set of Pacific Ocean Water samples after the initial volume of 20 L was reduced to around 5 L using a rotary evaporator.

## Acknowledgments

This work was supported by the Studienstiftung des deutschen Volkes and the Excellence Cluster Universe.

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# The impact of Fukushima accident in Sevilla (Spain) through the analysis of $^{129}\text{I}$ by Accelerator Mass Spectrometry

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**Keywords:** Fukushima accident, I-129, AMS

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$^{129}\text{I}$  is a long-lived radionuclide ( $T_{1/2} = 15.7 \times 10^6$  years) whose presence in nature nowadays is mainly due to artificial nuclear activities, specially discharges from nuclear reprocessing plants. Nuclear accidents like the one at Fukushima in 2011 have also injected amounts of  $^{129}\text{I}$  into the environment. As part of this radioisotope is emitted directly to the atmosphere, it can travel long distances, having an impact in zones far away from the origin. Thanks to its very long half-life,  $^{129}\text{I}$  can be analysed long time after the sample collection. However, very high sensitivity techniques are normally needed for its detection in environmental samples, for example Accelerator Mass Spectrometry (AMS).

In this work,  $^{129}\text{I}$  has been analysed in atmospheric samples taken in Sevilla (Spain) before, during and after the impact of Fukushima's discharged radioactivity arrived to the city. Three kinds of samples were prepared and analysed for  $^{129}\text{I}$  concentration measurement by AMS: rainwater, activated charcoal filters (for gaseous iodine adsorption) and polypropylene filters (for particulate iodine adsorption).

Samples were chemically processed and measured by AMS at the Centro Nacional de Aceleradores, Sevilla, Spain in a 1 MV compact AMS system.

The obtained results were compared to the previously measured in the same samples ( $^{131}\text{I}$  and  $^{137}\text{Cs}$ ) being found that the daily particulate  $^{129}\text{I}$  concentration profile between March 24<sup>th</sup> and April 10<sup>th</sup> 2011 is in very good agreement with the  $^{131}\text{I}$  concentration profile measured by gamma-ray spectrometry during these days in the same samples, showing that the origin of the analysed  $^{129}\text{I}$  was the accident in Fukushima on March 11<sup>th</sup>.

Maximum particulate  $^{129}\text{I}$  concentration of  $8.86 (0.56) \times 10^4$  at/m<sup>3</sup> was found on March 28<sup>th</sup>, while gaseous  $^{129}\text{I}$  concentrations had a maximum of  $7.7 (1.1) \times 10^5$  at/m<sup>3</sup>. Rainwater concentrations were typically in the order of  $10^8$  at/l, showing a maximum on April 3<sup>th</sup>.

In spite of the clear influence of Fukushima's accident,  $^{129}\text{I}$  concentrations before and after the impact of the accident in Sevilla were typically higher than the ones previously described. The  $^{129}\text{I}$  found normally in the atmosphere of Seville can be associated to the discharges of nuclear reprocessing plants, specially Sellafield and La Hague, and a detailed analysis of wind trajectories for the period analysed in this work suggest that there was a prevalence of western winds against northern ones, preventing the intrusion of  $^{129}\text{I}$  released by the nuclear reprocessing plants and giving evidence (in relation to  $^{129}\text{I}$ ) of the Fukushima impact in the city.

# The transport of close-in fallout plutonium in the Northwest Pacific Ocean : Tracing the water mass movement using $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in sediment

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Keywords: Sediment, NW Pacific,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ .

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The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio is a powerful tool to identify the sources of Pu and to be applied for tracing the water masses with the clarification of the transport processes in the ocean (Lee et al., 2013). In this study we demonstrate the characteristics of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in the sediment from the NW Pacific Ocean. The results we describe here provide a synthesis of information on the origin of Pu isotopes in the NW Pacific Ocean and contribute to a better understanding of transport processes in the region.

For this study  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in sediment collected from the Northwest (NW) Pacific Ocean from 1992 to 1997 were determined using ICP-sector field mass spectrometry (ICP-MS).

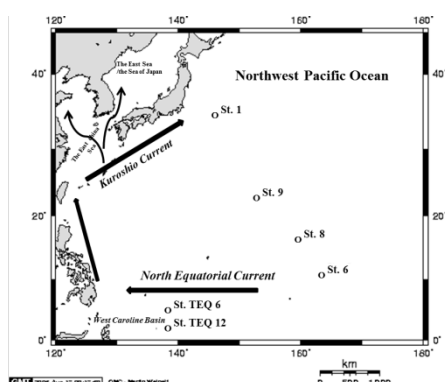


Figure 1 Sediment sampling stations from the Northwest Pacific Ocean

The results of atom ratio of  $^{240}\text{Pu}/^{239}\text{Pu}$  in the sediment in the NW Pacific Ocean and in the West Caroline Basin in the West Pacific Ocean are shown in Fig. 1. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in the surface sediment in the NW Pacific Ocean ranged from  $0.175 \pm 0.002$  to  $0.287 \pm 0.008$ . The atom ratios of  $^{240}\text{Pu}/^{239}\text{Pu}$  collected near the PPGs (at St. 8) was significantly higher than the global fallout ratio. However, in the sediment collected from St. 9 which is located in the north of PPGs, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio decreased. St. 1 located in the most northern part of NW Pacific in this study presented the lowest  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio ( $0.175 \pm 0.002$ ) which is consistent with that of the global fallout ratio. It is noted that the higher atom ratios than the global fallout ratio were found below 4 cm depth. In the West Caroline Basin in the West

Pacific Ocean, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios are  $0.217 \pm 0.012$  at St. TEQ 6 and  $0.183 \pm 0.011$  at St. TEQ 12, respectively. At St. TEQ 12 the atom ratio of  $^{240}\text{Pu}/^{239}\text{Pu}$  is consistent with that of the global fallout ratio in whole column while St. TEQ 6 showed high values.

Such elevated  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios indicate that the close-in fallout plutonium isotopes originating from the Pacific Proving Grounds (PPGs) due to the U.S. tests are prevailing in the sediment in the NW Pacific Ocean

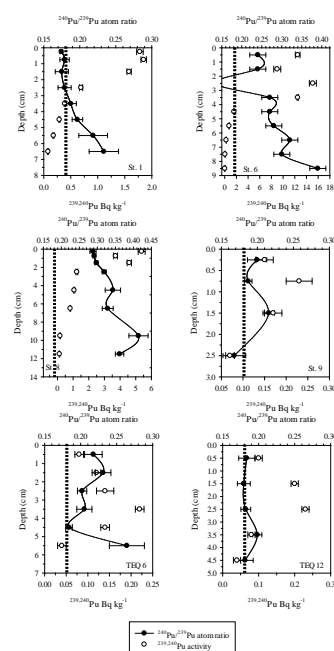


Figure 2. Depth distribution of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom in the NW Pacific Ocean and in the West Caroline Basin in the West Pacific Ocean

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# Man-made radionuclides in sediments of the Indonesian Throughflow

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Keywords: Am-241, Cs-137, Plutonium, Pacific Proving Grounds, Global fallout.

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The Indonesian Throughflow (ITF) as a part of the global ocean conveyor belt provides a low-latitude connection from the Pacific to the Indian Ocean. This complex array of passages within the Indonesian archipelago represents a pathway for radionuclide transport between both water bodies. Artificial radionuclides play a crucial role as tracers for transport processes in the Anthropocene and they offer important chronostratigraphic markers in high resolution recent sediment profiles.

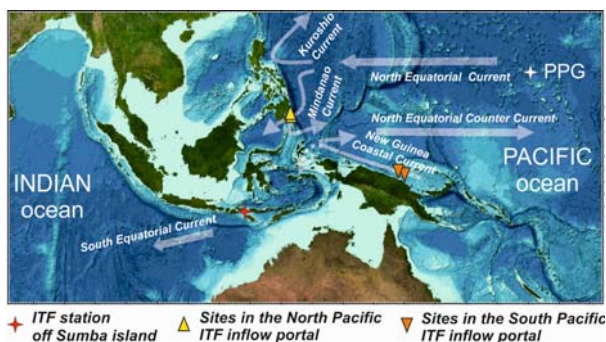


Figure 1. Sediment cores sampling sites (PPG: Pacific Proving Grounds)

In a previously studied deep sea site from off Sumba Island (Fig. 1), Indonesia, <sup>241</sup>Am and <sup>137</sup>Cs were analysed in sediment cores in order to support <sup>210</sup>Pb based chronology within a palaeoclimate interpretation of Australian-Indonesian summer monsoon (Steinke *et al.*, 2014). At this station unusually high <sup>241</sup>Am concentrations and high <sup>241</sup>Am/<sup>137</sup>Cs ratios were found. High <sup>241</sup>Am concentrations were reported previously in sediments from Cartier Trough, Timor Sea (Burns *et al.*, 2003).

The aim of present study is to examine artificial radionuclides in short sediment cores from 1) the ITF, 2) its North Pacific inflow portal, and 3) its South Pacific inflow portal. In addition to gamma emitters quantified by HPGe gamma spectrometry, Pu isotopes are studied by alpha spectrometry and mass spectrometric techniques (ICP-MS and AMS) to determine their inventories and ratios. The main objectives are to find out if enhanced Am concentrations are directly linked to high Pu contents, and to clarify the radionuclide sources based on their isotopic ratios. The contribution from the Pacific Proving Grounds (PPG) and global fallout, as well as some possible minor sources, will be quantified at all locations.

In some of the sediment gamma spectra, despite detectable <sup>241</sup>Am activities, <sup>137</sup>Cs was below decision

threshold (which was typically around 1 Bq·kg<sup>-1</sup> depending on sample mass and measurement time). In such cases summing spectra of several successive core slices enabled quantifying <sup>137</sup>Cs inventories.

Activities of <sup>241</sup>Am in sediment (to present date) were found as high as 12.6±1.1 Bq·kg<sup>-1</sup> in the northern Pacific inflow portal site, with <sup>241</sup>Am/<sup>137</sup>Cs activity ratios peaking at 5.2±1.4 at 10 cm depth below the sediment surface. The <sup>241</sup>Am/<sup>137</sup>Cs inventory ratio was found to be 3.4±0.2 at this site.

The study site off Sumba Island has a comparatively low, but still unusually high <sup>241</sup>Am/<sup>137</sup>Cs inventory ratio of 2.7±0.5. <sup>241</sup>Am activities exceeding those of <sup>137</sup>Cs were found also in sediment from the ITF South Pacific inflow portal.

Based on results of Pu isotope analysis we expect to test the hypothesis, that the source of the unusually high artificial isotope concentrations in this area is due to redistribution of local fallout from the PPG by ocean currents.

An additional implication of remarkably high <sup>241</sup>Am content in sediments of the ITF area appears to be its promising use as a supporting chronomarker in <sup>210</sup>Pb sediment chronologies. The most common artificial radionuclide used for this purpose is <sup>137</sup>Cs, however since its most prominent introduction to the environment during the period of atmospheric weapons testing its activity has decayed to 30% of its original level. The long-lived <sup>241</sup>Am (T<sub>1/2</sub>=432 yr) can be measured by HPGe gamma spectrometry with systems suitable for detection of low-energy gamma emitters, typical of those used for <sup>210</sup>Pb measurements. In order to use <sup>241</sup>Am as a chronomarker with high confidence, it is desirable to verify its origin (presumably as an in-situ decay product of <sup>241</sup>Pu) and therefore its chronological link.

## Acknowledgments

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# Application of $^{137}\text{Cs}$ , $^{14}\text{C}$ and Pu isotopes to trace pollutants in the Baltic Sea

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Keywords:  $^{137}\text{Cs}$ ,  $^{14}\text{C}$ , Pu isotopes, Baltic Sea.  
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$^{137}\text{Cs}$  and Pu isotope ratios in sediments,  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  of total organic carbon (TOC) as well as organic fractions of the sediments were applied to study sources and fate of organic carbon (OC) in the Baltic Sea including chemical warfare agent (CWA).

The compound-specific  $\delta^{13}\text{C}$  analysis, phospholipid-derived fatty acid biomarkers and a different end-member mixing model approach were used to estimate a relative contribution of the marine, terrestrial and fossil sources to organic OC in the sediments, to assess a possible effect of petroleum hydrocarbon contamination on radiocarbon signatures and to elucidate a probable leakage of CWA at the CWA dumpsite in the Gotland Deep.

Extraction of lipids from freeze-dried, homogenized surface sediments (0-3 cm) was performed according to published techniques (Zelles 1999; Tiwari et al. 2011).

The content and isotopic ratios of carbon in the samples were measured using a Thermo Scientific Delta V Advantage mass spectrometer coupled to a Flash EA 1112 elemental analyzer.

Measurements of  $\Delta^{14}\text{C}$  in sediments and in different classes of organic substances were carried out using a 1.0MV HVE Tandem AMS in the Department of Geosciences of the National Taiwan University.

$^{137}\text{Cs}$  activities were measured with HPGe detectors (GEM40P4-76, efficiency 40%, resolution 1.85 keV (FWHM) at 1.33 MeV and GX4018, resolution 1.8 keV/1.33 MeV and efficiency 42 %). Samples were ashed at 550° C and then dissolved in strong acids ( $\text{HNO}_3$ ,  $\text{HCl}$ ,  $\text{HF}$  and  $\text{HClO}_4$ ). The TOPO/cyclohexane extraction and radiochemical purification using UTEVA, TRU and TEVA resins (100 – 150  $\mu\text{m}$ ) were employed for separation Pu isotopes.  $^{242}\text{Pu}$  was used as yield tracers in the separation procedure. The overall recovery of  $^{242}\text{Pu}$  tracers was about 80% (Lujanienė, 2013).

The obtained data revealed a possible application of the Chernobyl-derived Pu to trace the pollutants of the terrestrial origin. The obtained results have indicated a different origin and behavior of lipids and TOC at the CWA dumpsite as compared to the area affected by the terrestrial-freshwater OC input. The high contribution of fossil sources at the CWA dumpsite (on the average 32%) as compared to other stations (on the average 14%) in the Baltic Sea may indicate a leakage from CWA. The  $\Delta^{14}\text{C}$  data of the TOC and total lipid extracts showed that recent sediments at the CWA dumpsite contained an excess of fossil carbon capable of influencing the radiocarbon dating at the site.

## Acknowledgments

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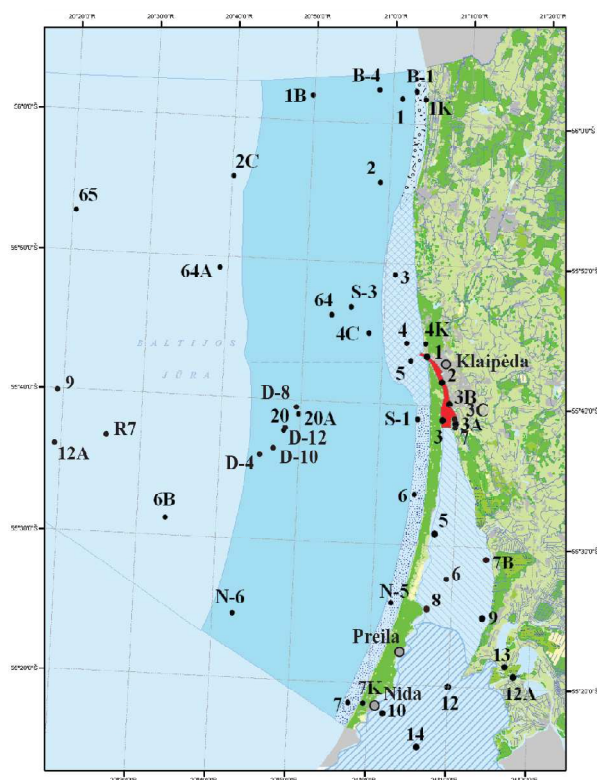


Figure 1. Sampling locations in the Baltic Sea and the Curonian Lagoon. Bottom sediment samples were collected during the sampling campaigns in the frame of the State monitoring and several other investigations in 1997 – 2014.



# Consequences on marine environment of a hypothetical accidental radioactive release in the Bay of Toulon: implementation of a marine dispersion model

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**Keywords:** hydro-sedimentary modelling, radionuclides marine dispersion, Bay of Toulon.

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In case of an accidental radionuclides release in marine environment, the contamination dispersion is controlled by many physical processes. Besides radioactive decay, radionuclides are dispersed by currents and might contaminate sediments and marine biota on a wide area. The environmental consequences and impacts on anthropic activities of a radioactive release are based on the dispersion of the contamination, which makes essential to foresee the contaminated areas.

Thus, the French Institute for Radiological Protection and Nuclear Safety (IRSN) develops post-accidental management tools to assess the radioecological consequences of this kind of contamination on marine environment. The prediction tool has to be based on a hydro-sedimentary model, since the future of a radioactive contamination is controlled by both hydrodynamic and sediment dynamic.

The Bay of Toulon (South of France) shelters the largest European nuclear harbour and this area has been chosen to set up a hydro-sedimentary model as a first study site. Located on the French Mediterranean coast, the Bay of Toulon is semi-enclosed and is divided in two basins connected by the channel entrance at the South of a large seawall (Fig.1). The question addressed by this study is hence: what would be the radionuclides future in case of an accidental release in the Bay of Toulon?

The coupling of the sedimentary and hydrodynamic MARS3D models allows simulating the sediments evolution. Since the tide amplitude is weak in the Mediterranean, waves and bottom currents have a large impact on re-suspension, transport and deposition. Previous works have aimed to validate the hydrodynamic model with current and waves model/measures comparisons and water exchange times assessments (Duffa *et al*, 2011, Dufresne *et al*, 2014).

The model distinguishes particles in classes and types, depending on their size and cohesive abilities. It calculates the transport within the water column and the suspended-sediment concentration fluxes to simulate the exchanges due to erosion and deposition near the bottom. Dissolve/particulate proportion for radionuclides is calculated with distribution coefficient ( $K_d$ ), based on IAEA (2004).

Hydrodynamic of the Bay and water mass exchanges through the channel have been studied. Results are highly linked to the atmospheric conditions and the water exchange times of the Little Bay (LB, see Fig.1) range from one to six days, depending on wind conditions. Recorded data in the Las River that flows

into the LB led to the description and the estimation of the catchment yields to the Bay, poorly studied before. With one ton of particulate matter annually discharged by the Las River, the LB appears to be a sedimentation area.

As an example of application, a hypothetical discharge of  $10^9$  Bq of  $^{137}$ caesium is numerically added in the model at the military harbour location (grey dot in Fig.1). Results show that the dissolve contamination concentrations are linked to the wind-induced currents and to the water exchanges at the channel. Windy days lead to a faster decrease of dissolve  $^{137}$ Cs activity in the LB. For this specific scenario, 90% of the initial  $^{137}$ Cs is out of the LB one week after the input. After 2.5 months, 96% of the initial input is out of the LB while 4% is trapped in its bottom sediment.

Since the bottom sediment is covered by mud and the hydrodynamic is reduced by a seawall, the LB appears to be a trapping zone and hence a sensitive area in case of accidental release of radionuclides.

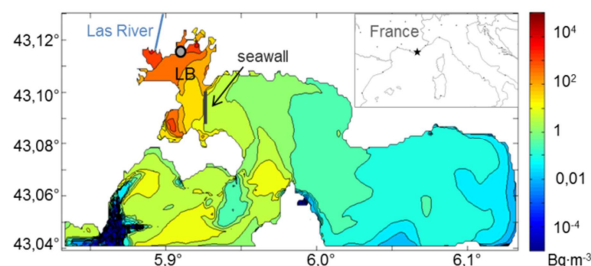


Fig.1: Simulated  $^{137}\text{Cs}$  activity ( $\text{Bq}\cdot\text{m}^{-3}$ ) in bottom sediment 30 days after the input (grey dot) and location of the Bay of Toulon (black star).

## Acknowledgments

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# Recent coastal sedimentations rates from NE Mediterranean using $^{210}\text{Pb}_{\text{ex}}$ and $^{137}\text{Cs}$

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**Keywords:** Sedimentation rate, Fallout radionuclides,  $\gamma$ -spectrometry

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Most radionuclides entering the marine environment accumulate at the coastal zone and therefore their concentrations and distribution in the sediment are of great interest. Coastal and marine sediment's radioactivity is related to marine pollution and sedimentation processes, factors important for better management and protection of the marine resources. Especially in the case of semi-closed and closed aquatic ecosystems, which usually act as dump of leach materials due to human activities around the catchment area, radiochronology of marine sediments is particularly important as it provides information not only on geological phenomena (e.g. earthquakes, volcanic activity) but also on physicochemical processes (e.g. bioturbation, shallow deposits diagenesis, chronological determination of industrial pollution) and site specific macroscopic characteristics (e.g. radionuclides' diffusion coefficient in the sediment, vertical dispersion velocity).

In this frame, the knowledge of radionuclides' activity concentration vertical profiles can give valuable information. The main radiotracers widely used in the investigation of sedimentation processes are the so called Fallout Radio-Nuclides (FRN) and in particular: the excess or unsupported  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_{\text{ex}}$ ), which is not in equilibrium with the parent  $^{226}\text{Ra}$ , and the anthropogenic  $^{137}\text{Cs}$  (Marbit *et al*, 2008).

Table 1. Core sediments' sampling positions.

Location	Date	Station	Depth (m)	Description
Thessaloniki Gulf	2005	AM(2,4,5,7,10)	5-14	open gulf
Litohoro Bay	2009	LT-10	45	estuary
Amvrakikos Gulf	2007	13A	15	strait
		13B	40	closed gulf
Uluabat Gölü	2007	ΠNT-KI-07	3	lake
Gulf of Corinth	2007	COR2b	780	rift slope
Elefsis Gulf	2012	S2	33	semiclosed gulf

In this work, core sediments samples collected from selected sites of the NW Mediterranean Sea covering different costal environments (Table 1) have been analyzed for their radioactivity. Natural and anthropogenic radionuclides were measured using HPGe detectors, while the vertical distributions of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  at the sediment cores were used to determine recent sedimentation rates. The study involves new measurements and reanalysis of previous data (Tsabaris *et al*, 2010; Papageorgiou *et al*, 2011; Tsabaris *et al*, 2012), covering a range of different aquatic environments and a wide costal area.

From the  $^{210}\text{Pb}_{\text{ex}}$  activity concentration vertical profiles and according Sanchez-Cabeza, J.A. and Ruiz-

Fernandez (2012) formalism, the sedimentation rates were determined at the sampling stations applying three different calculation methods: the Constant Flux-Constant Sedimentation (CFCS), the Constant Rate of Supply (CRS), the Periodic Flux (PF) and Constant Initial Concentration (CIC) models. The mean sedimentation rates after 1963 have been also estimated from the peaks of  $^{137}\text{Cs}$  vertical profiles (Tsabaris *et al*, 2012).

Summarizing the results, increased sedimentation rates were observed in all examined cases with values varying from 0.18 to 0.55 cm/y. This can be attributed to the selection of the sampling stations representing, each for different reason, particular dynamic aquatic environments. From the data analysis it was also clear that the application of multiple methods is complimentary and that the use of different approaches and tracers can give more reliable and robust estimations.

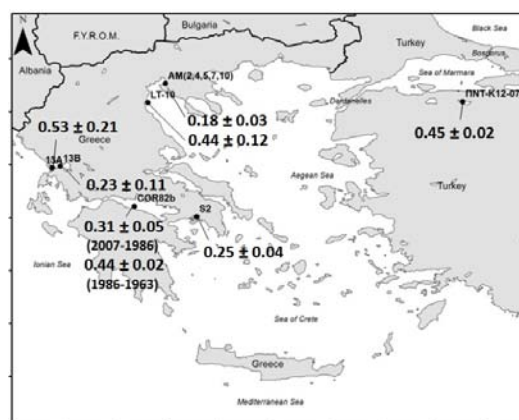


Figure 1. Estimated sedimentation rates (in cm/y).

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# Disequilibrium in the uranium and actinium series in oil scale samples

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Keywords: oil exploration, <sup>235,238</sup>U, <sup>232</sup>Th  
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Radiation in the oil exploration industry was first identified as early as 1906, a short ten years after Becquerel discovered radioactivity. In the 1980's <sup>226</sup>Ra began to be noticed when scrap metal dealers would detect unacceptably high levels of radiation from oil-field piping. TENORM will develop in high concentrations in by-product oil and gas waste streams. The NORM will chemically separate from other piped material in the process of the extraction of oil, resulting in high concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>210</sup>Pb and other radioisotopes in a densely caked layer on the inner surfaces of the piping, which is called scale. Sludge is often part of the process. The activity of the <sup>226</sup>Ra from TENORM ranges from 185 to several tens of thousands Bq/kg of sample. Often overlooked are the activity levels of <sup>226</sup>Ra and <sup>210</sup>Pb. The environmental pathways of uranium, thorium and lead in the oil fields are different and thus disequilibrium occurs in the scale. Figure 1 shows the rich concentrations of the TENORM in a typical sludge sample from oil processing.

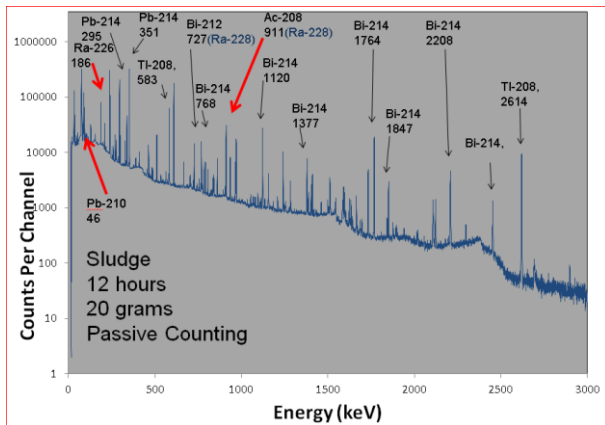


Figure 1 Typical gamma-ray spectrum of sludge in oil exploration by-product.

Besides typical gamma-ray counting self-attenuation of the photons, especially the 46 keV gamma-ray from <sup>210</sup>Pb must be taken into consideration. Scale has an abundance of strontium, calcium and barium precipitates in its matrix since they belong to the same Group II in the periodic table. Figure 2 the % Transmission vs Energy in a scale sample. We will present detailed characterization of the disequilibrium in a typical scale sample as well as using neutron activation analysis to determine <sup>238</sup>U and <sup>232</sup>Th.

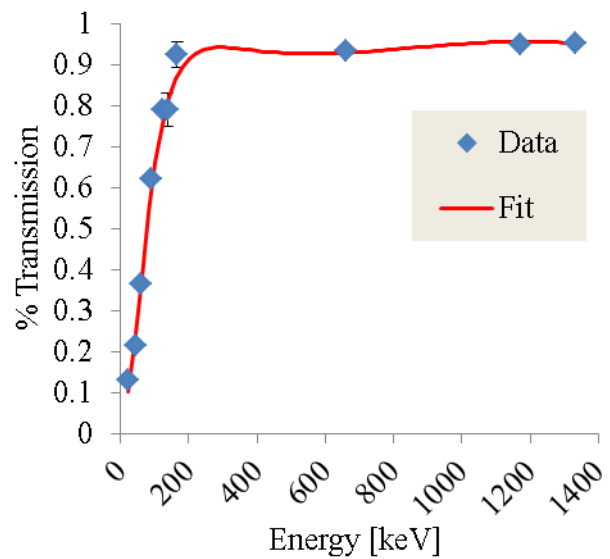


Figure 2 % Transmission vs Energy in a scale sample.



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## Accelerator mass spectrometry laboratory at the Comenius University in Bratislava: first results

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**Keywords:** accelerator mass spectrometry, Pelletron, tandem accelerator, MC-SNICS ion source, ion analyser.

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Accelerator Mass Spectrometry (AMS) laboratory established recently at the Centre for Nuclear and Accelerator Technologies (CENTA) at the Comenius University in Bratislava consisting of a MC-SNICS ion source for solid targets, 3 MV Pelletron tandem accelerator, and an analyzer of accelerated ions (Povinec et al., 2015a, Ješkovský et al., 2015) will be presented. A preparation of targets for <sup>14</sup>C and <sup>129</sup>I AMS measurements will be described shortly as well.

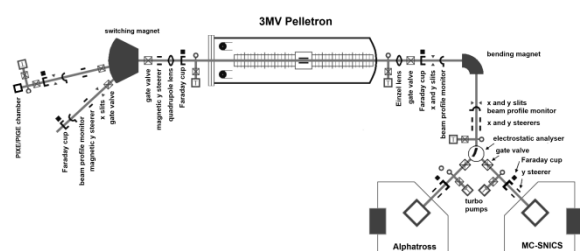


Figure 1. Floor scheme of the present structure of the tandem laboratory.

The first AMS measurements included <sup>10</sup>Be using a switching magnet at 45°deg and an ionization chamber with silicon nitride stack absorber for <sup>10</sup>B isobar suppression. The obtained detection limit for <sup>10</sup>Be/<sup>9</sup>Be mass ratio was around 10<sup>-12</sup>. <sup>14</sup>C analysis of tree-ring samples (Povinec et al., 2005b) from the vicinity of the

Jaslovské Bohunice nuclear power plant (NPP), and from Bratislava will also be discussed.

A development of AMS technique for potassium, uranium and thorium analysis in radiopure materials required for ultra-low background underground experiments (Povinec et al., 2015c) will be briefly mentioned as well.

### Acknowledgments

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## Measurement by gamma ray spectrometry in gamma-gamma coincidence.

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**Keywords:** *Gamma-ray spectrometry, gamma-gamma coincidence.*

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The laboratory of environmental radioactivity measurement performs around 2000 measurements per year by low level gamma ray spectrometry, either in the framework of the environmental surveillance in France handled by the IRSN, or for research or expertise purpose in the radioecology domain. In both cases the anthropogenic radioactivity in environment samples is measured at trace levels and metrology developments are continuously necessary in order to deal with these very low levels.

The latest development is the gamma-gamma coincidence measurement based on the use of two germanium detectors to detect one photon in each detector.

This method was firstly implemented on an anti-Compton system, consisting of a germanium (HPGe) detector surrounded by two sodium iodide scintillators (NaI(Tl)): an annulus and a plug to cover around  $4\pi$ ). The digital acquisition electronics provides a list mode file with energy and time stamp information for each detected event. An algorithm was developed to plot spectra (classic, in coincidence or in anti-coincidence) or the coincidence matrix (energy loss in the NaI detector versus energy loss in the HPGe). The algorithm searches and counts the coincidence events of interest in the matrix around their theoretical location and the corresponding activity is determined by way of a calibration factor. It was obtained by the experimental measurement of standard sources containing for instance <sup>60</sup>Co or <sup>88</sup>Y and by Monte Carlo simulations using MCNP\_CP (Berlizov,2013). Both calibration coefficients are perfectly in agreement and the method can be extended to all radionuclides: <sup>134</sup>Cs, <sup>152</sup>Eu, <sup>108m</sup>Ag...

This analysis method was therefore used on a new versatile system developed and optimized in terms of efficiency, low background and compactness: it consists of two HPGe detectors face to face surrounded by a NaI(Tl) scintillator. The whole system is surrounded by a 5 cm lead shield. This new system is installed in a 20 m<sup>2</sup> shielded

room (10 cm lead, 5 mm copper) located in the second basement of the laboratory under a slab of 3 meters of borated concrete.

Thanks to the analysis code of the list mode file, a measurement can be analysed at once in the following different ways:

1. **Classic Ge spectrum for each Ge detector.**
2. **Summation of spectra of both detectors.** The efficiency increase leads to decrease uncertainties and detection limits and counting time for single emitters.
3. **Anti-cosmic spectrum.** The cosmic ray induced background decrease leads to a better determination of <sup>137</sup>Cs in aerosol filters for instance.
4. **Anti-Compton spectrum.** The Compton continuum decrease leads to a better determination of single emitters (with energy lower than 1460 keV) in biological matrices containing much <sup>40</sup>K.
5. **Coincidence Ge-Ge matrix of Ge spectrum in coincidence.** This is the most powerful tool for all the coincident emitters due to a drastically decreased background. Moreover some radionuclides have many coincident emissions and all the data can be accumulated (for instance, <sup>108m</sup>Ag has 6 usable coincidences) to make the determination more accurate.

The gamma-gamma coincidence techniques used on the anti-Compton system divides detection limits by a factor of 10 (It shows also that the calibration can be done by MCNP\_CP simulations). First results on the new Ge-Ge-NaI system show dramatically reduced noise in matrices and spectra and all the possibilities with only one measurement.

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# Monte Carlo simulation of probe response to calibration measurements, Straz pod Ralskem calibration facility, Czech Republic

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Keywords: MCNPX, calibration facility, portable gamma-spectrometry  
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DIAMO, state enterprise, Straz pod Ralskem is the only calibration facility for portable gamma-ray spectrometers in the Czech Republic. The calibration facility was constructed by the Czechoslovak Uranium Industry in 1975, originally in Bratkovice near Píbram. It serves for calibration of portable, car-borne, airborne and logging gamma-ray spectrometers used in geological mapping, uranium exploration and environmental studies. The facility was moved to Straz pod Ralskem in 2009. Portable ground gamma spectrometers are calibrated using 4 cylindrical concrete pads with diameter 1.9 m and thickness 0.62 m. There are standards of K, U, Th and background standard. (Rojko et al., 1975)

For precise measurements, which are required for instrument calibration, the effective concentration of elements in the pads needs to be known precisely. In the same time the other possible sources of gamma radiation which may contribute to the spectra needs to be considered. The concentrations, which are used for calibrations in Straz pod Ralskem calibration facility, were determined using laboratory gamma spectrometry and verified using a pair of transportable gamma spectrometers- GR-320 and GS-256). The influence of other sources was analyzed on the basis of collimated measurement. (Matolín et al., 2012)

The main aim of this work is to verify the concentrations of calibration pads which are used in Straz pod Ralskem. The detailed model of whole facility for Monte Carlo simulation (MCNPX) was created for the needs of this work. The responses of NaI(Tl) 3x3" (GF Instruments- Gamma Surveyor) detector were calculated for all the possible sources of radiation. The considered sources were: calibration pads, the floor, the walls and also the air in the room. Simulated radionuclides were <sup>40</sup>K, <sup>214</sup>Bi (<sup>238</sup>U decay chain daughter product), <sup>208</sup>Tl (<sup>232</sup>Th decay chain). To verify the results of our model the series of measurements were performed using the same probe which was previously simulated.

Measurements of radon concentration in air and also a concentration of radon daughter products were also performed. These data serve to evaluate the contribution of <sup>214</sup>Bi present in air to measured spectra. Samples of building materials were collected for

laboratory semiconductor gamma spectrometry. The concentrations of K, U and Th, measured in these samples, were included in the prepared model.

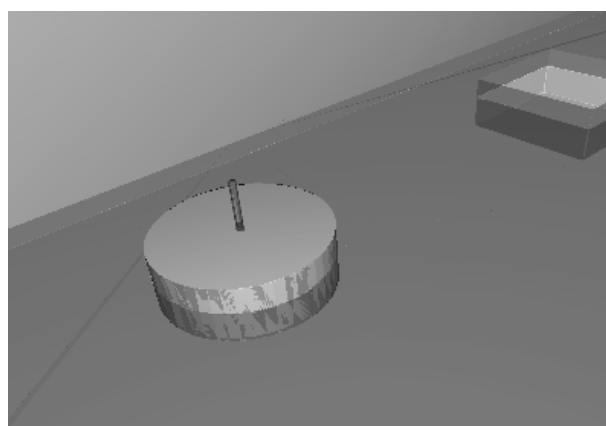


Figure 1: Visualization of NaI(Tl) probe placed on U concrete calibration pad

Our work presents a comparison of calculated and measured results and discusses the accuracy of data which are provided for making calibrations of portable spectrometers in Straz pod Ralskem calibration facility. The work also discusses some limitations of Monte Carlo simulations and their usability for simulation of spectrometer response to calibration measurement.

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# Evaluation of the environmental impact of the European Spallation Source facility

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**Keywords:** spallation source, source term, dose factors, effective dose.  
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The European Spallation Source (ESS) is the European common effort in designing and building a next generation large-scale user facility for studies of the structure and dynamics of materials. The proposed schematic layout of the ESS facility is based on a linear driver (linac) directing the proton beam (5 MW of 2 GeV) of 2.8 ms long pulses with a 20 Hz on a tungsten target where neutrons are produced via spallation reactions. Further the neutrons will be moderated to thermal and subthermal energies in a couple of 22 moderators placed around the target and guided to the scattering instruments, mainly for neutron scattering research.

This document assesses the radiological consequences from potential radioactive waste discharges arising from ESS and straight radiation into the environment during the routine operation of the facility. These discharges are in form of gaseous discharges to the atmosphere from a stack, aqueous discharges to public sewer and migration of contaminant with the groundwater.

The source term (ST) of atmospheric releases was separated into two distinct release operations (Ene, 2013): i) on-line emissions, and ii) emissions resulting from processing. The continuously release from the accelerator tunnel (AT) was derived based on the activation calculations of the air and the designed ventilation rate of 4320 m<sup>3</sup>/hr. Releasing from the target station (TS) is mainly due to the leakage (0.1% per day) from the helium cooling loop of the target, see Figure 1. The ST for TS was derived using the calculated radioactive inventory of the target, assumed release fractions within the cooling loop and further effectiveness of the purification system for removal of the tritium and halogens.

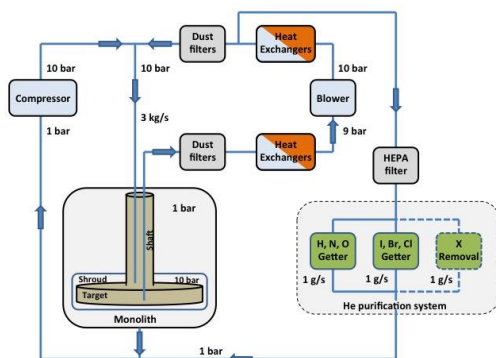


Figure 1. Schematic view of the Helium cooling loop.

ST of processing operations accounts for: i) dismantling the target within the hot cell, and ii) cementation of liquids within the waste facility. ST for the groundwater pathway was derived using the calculated activity of the soil around the accelerator tunnel, for a beam loss of 1W/m. Discharges to the sewage plant were assessed considering only tritium into the wastewater.

The PREDO platform (Sundell-Bergman S et al., 2014) was used to carry out radioecological and dose assessment models. Main parameters and assumptions used to define dispersion were based on Swedish Regulator requirement (SSM, 2009). Two reference groups were defined: an existing farm family exposed due to agriculture, and a hypothetical group (HP) exposed due to the presence. Results of calculations lead to a total effective dose of 28  $\mu$ Sv/y. The most demanding dosing operations are given in the Figure 1.

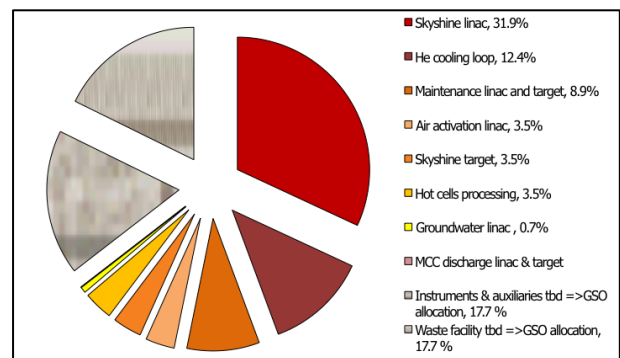


Figure 1. Main components of routine releases

Conservative assumptions were made for occupancy and intake data used. Estimated annual effective doses for three age groups and each pathway are breakdown in the paper. The maximum dose estimate for the farm is about 11  $\mu$ Sv/year due mainly to <sup>32,33</sup>P and <sup>35</sup>S. In case of the HP the largest annual doses of about 1  $\mu$ Sv/year arises from the nuclides <sup>11</sup>C, <sup>13</sup>N and <sup>3</sup>H. Impact due to water discharges was found to be negligible.

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# A new generation of detectors for environmental radiation monitoring

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**Keywords:** spectrometer, early warning networks, environmental radiation monitoring  
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In the case of a major radiological emergency with a release of radioactive material, early and reliable information about the activity concentrations especially in urban areas is of key importance for appropriate governmental decisions on countermeasures to limit economical and social consequences. The follow-up costs in case of a nuclear power plant accident like e.g. those occurred in Chernobyl and Fukushima may exceed hundred billions of dollars. As a direct consequence of the emergency in Chernobyl in 1986, all European countries have installed radiological early warning network systems. Most of the measuring stations supply only dosimetric information, as they are mostly equipped with dosimeters based on gas filled detectors.

In a number of articles, like that of Toivonen *et al.* (2008), it was demonstrated, that the use of spectrometry systems can give both information on the contamination levels (nuclide specific information) and on ambient dose equivalent rate values. In order to retrieve and calculate dose rate values from energy spectra several methods exist. Some basic approaches are described and compared with each other in Dombrowski (2014). Advantageous is a method which allows the deduction of ambient dose equivalent rate values from spectra by using energy-dependent conversion coefficients (without the need of a deconvolution). The practical application of this method is demonstrated in this article using the example of the characterisation of four different detector systems: a semiconductor detector (CdZnTe) and three scintillation detectors (LaBr<sub>3</sub>, CeBr<sub>3</sub> and SrI<sub>2</sub>). Results of the angular and the energy dependence of the detector's response function will be shown and conversion coefficients from count rate to ambient dose equivalent rate will be given.

All detector systems are mounted in a weather-protected housing, so that they are suitable for outdoor use. The out-door use has been tested on the PTB reference measuring site for ambient radiation.

The method to use spectrometers for dose rate measurements is verified by irradiations in the underground PTB low-dose rate calibration facility, which is traceable to primary PTB standards. Especially measurements in the photon field of a sealed <sup>226</sup>Ra source, which emits a  $\gamma$ -ray spectrum comparable to that found in the natural environment, can be regarded as a meaningful verification.

The spectrometers mentioned above were exposed to Rn-progeny atmospheres in a Rn-chamber at PTB. This chamber has a volume of about 20 m<sup>3</sup> in

order to minimize the influence of progeny attached to the chamber's walls. The experimental conditions in this chamber are very well known and therefore ideally suited to study the influence of radon progeny on spectrometers and dosimeters. It is possible to compare the dose rates derived from the spectrometry systems with those directly measured with various dosimetry systems. As an example, the spectrum of the Rn-progeny measured in the PTB Rn-chamber with a CeBr<sub>3</sub> spectrometer is shown in the Figure.

The suitability of novel types of spectrometers for environmental radiation measurements will be discussed.

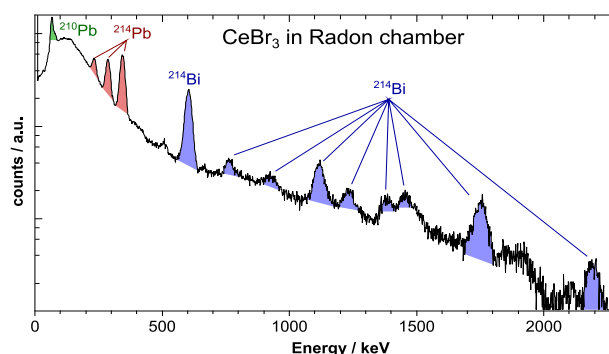


Figure:  $\gamma$ -ray spectrum of <sup>222</sup>Rn-progeny measured with a CeBr<sub>3</sub> detector. From this spectrum the ambient dose equivalent rate was derived using appropriate conversion factors.

## Acknowledgments

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# Use of spectroscopic and microscopic methods to reveal the evidence of U(VI) sorption on *Acidovorax facilis* isolated from subsurface environments

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**Keywords:** *Acidovorax facilis*, uranium, TRLFS, TEM/EELS.

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For bioremediation of uranium contaminated environments from activities such as uranium mining and uranium processing, microorganisms could be important due to their ability to immobilize radionuclides and heavy metals. Since the main public concern is the possibility of radionuclide escaping and migrating into groundwater, there is an intense interest in the development of effective remediation methods. The aim is to improve bioremediation strategies, based on a better understanding of binding mechanisms on the molecular level.

For our studies we used *Acidovorax facilis* (formerly *Pseudomonas facilis*), an aerobic Gram-negative Betaproteobacteria, which is commonly found in soil. Experiments were performed in batch cultures under aerobic conditions at 25 °C using nutrient broth. The cells were grown to an optical density (OD<sub>600</sub>) of around 1.5. For U(VI) biosorption experiments the cultures were washed 2 times with tap water and then resuspended in tap water. After that UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> was added to the solution to achieve an initial uranium concentration of 0.05 and 0.1 M, respectively, at a neutral pH range. The duration of the sorption experiments were limited to 48 h. As a response to uranium stress *Acidovorax facilis* were forming extracellular polymeric substances (EPS) resulting in the formation of cell agglomerates. For separating the EPS from the bacteria, the cell agglomerates were ultra-centrifuged (40.000 x g) for 2 h at 10 °C. The cell pellet was used for time-resolved laser fluorescence spectroscopy (TRLFS). The U(VI) luminescence at 274 K was measured after excitation with laser pulses at 266 nm and with an average pulse energy of 300 µJ. The emitted fluorescence light of the cell pellet was recorded using an iHR550 spectrograph and an ICCD camera in the 370 – 670-nm wavelength range by averaging 100 laser pulses and using a gate of 2000 µs. The data were analysed using Origin software, version 8.6, including the Peak Fit module, version 4.0.

The measured emission spectrum of the pellet is characterized by four emission bands, as shown in the luminescence spectrum (Fig. 1). Their peak maxima were observed at 497.8, 519.5, 544.1 and 568.6 nm ± 0.5 nm. In addition, the spectra of the Uranyl-lipopolysaccharide-complexes R-O-PO<sub>3</sub>-UO<sub>2</sub><sup>2+</sup> and [R-O-PO<sub>3</sub>]<sub>2</sub>-UO<sub>2</sub><sup>2+</sup>, reported by Barkleit et al. (2008), were used for comparison. The reference spectra display band positions at 498.1, 519.6, 542.9 and 567.5 nm for pH 4 as well as 499.7, 521.0, 544.3 and 568.9 nm for pH 7. They show only a small deviation from those

observed in our studies. Hence, it can be concluded that phosphoryl groups are the main binding sites for uranyl, located in the lipopolysaccharide (LPS) unit in the outer membrane by Gram-negative *Acidovorax facilis* cells.

Figure 1. Luminescence spectrum of *Acidovorax facilis* cells at 274 K exposed to U, in comparison with reference spectra of Uranyl-LPS-complexes (Barkleit et al., 2008).

After the U(VI) biosorption experiments, *Acidovorax facilis* cells were prepared for Energy-filtered transmission electron microscopy (EF-TEM) and electron energy-loss spectroscopy (EELS) by following the routine embedding protocol as described by Lünsdorf et al. (2001). The results provide microscopically and spectroscopically evidence of Uranium sorbed at the outer membrane of *Acidovorax facilis* cells by showing high electron density and U ionization intensity peaks. The results support the TRLFS measurements and contribute to a better understanding of the binding mechanisms of U(VI) on *Acidovorax facilis* cells.

## Acknowledgments

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# Comparison of adsorption properties of Cs on different types of activated carbon

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**Keywords:** Activated carbon, adsorption, caesium, water remediation

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Cs-137 ( $T_{1/2} \approx 30$  years) is an important long-term contributor to environmental contamination. It is released into the biosphere by nuclear weapons testing or reactor accidents (Whicker et al., 2007). For example, the release by the accident at the Fukushima Daiichi nuclear power plant raised the Cs-137 concentrations in the local groundwater to concentrations above the drinking water limit in downstream areas like the Fukushima port (TEPCO, 2013). Cs salts are very soluble in water, but strongly bind to soils and minerals (Public Health Service, 2004). These properties are used to investigate the removal of Cs from water using a wide range of low-cost adsorbents (Li et al., 2014).

In order to remove (radio-)caesium from water, adsorption on activated carbon (AC) is suggested as an economical and straightforward method to concentrate low levels of Cs on a compact adsorbent. Considering the high pH point of zero charge of several AC's, adsorption should be performed at high pH (Hanafi, 2010). The goal of this work is to determine the best adsorption conditions for removal of low levels of Cs from radioactive wastewater, using AC. In this experiment, AC's from brewer's spent grain (BSG) are compared to two commercially available industrial AC's. The BSG AC's are produced in a tube reactor using steam activation (Vanreppelen et al., 2014).

A 1000 ppm Cs ( $\sigma = 29$  barn) standard solution (VWR CertiPur, traceable to NIST) was irradiated in the neutron flux of the BR-1 reactor at SCK-CEN Mol to obtain a Cs-134 solution. From the mother solution, three dilutions with different pH (7, 10 and 12) were prepared, adjusting the pH with ammonia. Dilution factors and concentrations were calculated by measuring the samples in a NaI well detector (nearly 100% efficiency for Cs-134) and in an ionisation chamber. Cs-134 serves as a tracer for the total amount of Cs in the solution and its activity can be correlated to the concentration.

Adsorption experiments are carried out at 3 different pH's using 10 different AC's: 2 commercially available AC's (Norit G1240, Filtrasorb F400), 3 AC's from BSG, and the same 5 AC's previously loaded with a small amount (0.5 %) of Prussian Blue (PB) prior to the adsorption experiment in order to further enhance Cs adsorption. The amount of PB on the AC's is determined by calculating the difference between the iron concentrations (measured via ICP-AES) of a PB solution before and after adsorption of PB on AC. Approximately 25-30 mg of AC is put into contact with 9 g of the active Cs solution ( $\pm 1.2$  ppm Cs; 0.6 Bq/g) and the total activity is determined by measuring the tubes in the NaI well. These tubes are shaken for 36 hours. The solution is

filtered using plastic funnels and Whatman ashless filters to separate the AC from the solution. The emptied tubes, the filters with AC and the tubes with the collected solutions are measured in the NaI well. From the measurement, the fraction of Cs-134 adsorbed on the AC and the fraction remaining in the solution are calculated.

As shown in figure 1, adsorption percentages between 15 and 20 % are reached for both commercially available AC's and lab-scale AC's from BSG at pH 12. The highest adsorption rate was reached using the AC from BSG (activated with steam at 800 °C for 45 minutes with 10 ml of water (ACBSG07)), with an adsorption capacity of 18,6 % (no loading) and 20,3% (loaded with PB).

Adsorption of Cs at low levels in water is feasible using AC as adsorbent. Modification of the AC for optimal adsorption of Cs can be considered, as well as purification of Cs contaminated water in sequential adsorption stages.

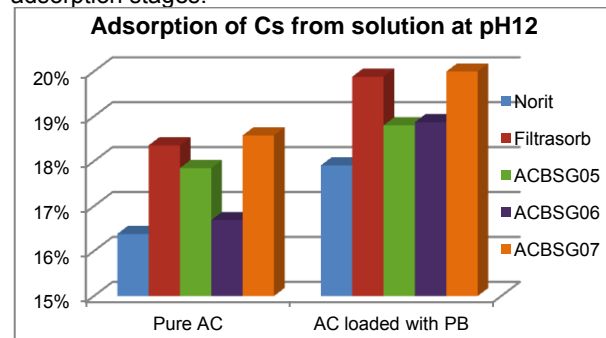


Figure 1. Adsorption percentage at pH 12.

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# Affinity Capillary Electrophoresis in Studying the Complex Equilibriums of Radionuclides in Aqueous Solution

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Radionuclide migration in the environmental systems is an important concern. Knowledge of its transport through the environment is crucial not only for fundamental geochemistry but for assessing the risk posed by long-term storage of nuclear waste. The problem of proliferation of uranium contamination after widespread use of depleted uranium in certain world regions is also important (Oliver et al., 2008).

Among a variety of possible geochemical processes, complex formation with inorganic and organic species present in water plays an important role in the eventual dispersion of radionuclides (Hummel et al., 2005). The formation of such complexes causes significant changes in mobility of waste materials. The overall change of mobility will be defined by the stability of the complexes; therefore, to predict dispersion of radioactive material we need to know thermodynamic parameters (such as stability constants,  $\Delta G$ ,  $\Delta H$ , and  $\Delta S$ ) of their complexes.

Affinity capillary electrophoresis (ACE) is used for examining binding parameters between biological species in past twenty years (Chu et al., 1995) (Jiang and Armstrong, 2010). The method is based on change in the electrophoretic mobility of detected species due to the interaction with other species present in electrolyte.

This paper is devoted to the use of ACE for study of radionuclide equilibriums in aqueous solutions. Special attention is paid to methodological development of ACE for the determination of:

- number of species involved in equilibrium;
- species constituents (number of ligands, protonated, deprotonated);
- stability constant values of complex formed;
- $\Delta G$ ,  $\Delta H$ ,  $\Delta S$  (influence of temperature).

The case of uranium (U(VI)) and selenium (Se(IV) and Se(VI)) is used for illustration (Sladkov, 2010) (Sladkov, 2013). These species are chemically and radiologically toxic.  $^{79}\text{Se}$  is a long-lived fission

product can be created from the thermal neutron fission of  $^{235}\text{U}$  in nuclear fuel plants. The interaction of U(VI) with acetic acid, as a primordial of complex organic molecules, is also considered (Sladkov, 2014). Acetic acid can represent the most simple model substances for humic acids which can occur as humic acid building blocks and simulate their structure and functionality (Sachs et al., 2011).

Thus, it is demonstrated, that the use of ACE can be fruitful for study of complex equilibriums of radionuclides and give valuable information for further modelling of behaviour of hazardous materials under environmental conditions. Moreover, when we work with hazardous materials, the following features of ACE are especially valuable: small sample amount requirements, low reagent consumption, relatively low cost and aptitude for automation and set-up in a glove box.

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# Retention efficacy and release of radioiodine in fumehoods

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Keywords: radioiodine, environmental radioactivity, fumehood, retention efficacy  
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The release of radioiodine to the environment has to be reduced to regulatory limits. Details concerning the construction and test of fumehoods are specified by a European norm (EN 14175). The method to determine the outbreak of hazardous gaseous substances is based on the detection of sulphur hexafluoride ( $\text{SF}_6$ ). The detection limit of this test is  $4.5 \cdot 10^{-7} \text{ mol / m}^3$  in exhaust air. This method may not be appropriate for determination of exhaust air containing radioactive substances, especially non-carrier-added radionuclides. This detection limit would represent a very high activity in the region of  $0.27 \text{ TBq / m}^3$ . Therefore, we developed a test using a filter-system (Fig. 1, Fig. 2) for selective adsorption of chemically different radioiodine species as described by Schomäcker et al. [Schomäcker (2011)] where the calibration procedure is also described.

We investigated the retention efficacy for two fumehoods (A1, A2) with a charcoal filter combination for which the routine test with  $\text{SF}_6$  showed no defects. Tests were performed at different sites. We performed the following experimental set-ups:

Set-up 1. Measurement of radioiodine in the vicinity of the fumehood (Fig. 1). Defined chemical species and amounts of radioiodine were released in the fumehood and measured in close proximity.

Set-up 2. Decontamination capacity of the fumehood (Fig. 2). With this test the activity in the exhaust air immediately after passage of the filter was measured.

Set-up 3. Radioiodine concentration in the room air at different positions where the air might be discharged into the environment.



Fig. 1 Set-up 1



Fig. 2 Set-up 2

The radioiodine depletion by fumehood ventilation was found to be 1:24 (A1) and 1:120 (A2). The fraction of organically bound iodine species varies when the fumehood ventilation is switched on due to the high volatility of monoiodoethane. In the room air we measured more organically bound iodine ( $30 \text{ C}_2\text{H}_5\text{I} : \text{I}_2$ ).

## 1. Outbreak

		Fraction of released radioiodine (% per $\text{m}^3$ )	
		A1	A2
1cm	Left	0.0018	0.000115
	Middle	0.00015	0.000145
	Right	0.0013	0.000269
6 cm	Left	0.0004	n.d.
	Middle	0.000069	n.d.
	Right	0.000049	n.d.

## 2. Decontamination capacity

		Fraction of released radioiodine (% per $\text{m}^3$ )	
		0.0013	0.000094

## 3. Room air concentration

		Fraction of released radioiodine (% per $\text{m}^3$ )	
	Pos. 1	$3.6 \cdot 10^{-7}$	n.d.
	Pos. 2	$8.6 \cdot 10^{-6}$	n.d.

Tab. 1 Measurements in the vicinity of the fumehood, after filtration and in the room air

The method of testing the retention of radioiodine in fumehoods presented in this work is more effective than the test with  $\text{SF}_6$ . The respective European norm is therefore not suitable for radioactivity in fumehoods. For one fumehood (A1) the retention was insufficient and measurable amounts of radioiodine were found in the laboratory.

The German legislation defines an exemption level of  $5 \text{ Bq/m}^3$ . Taking this level into account the maximum activity that can be handled in this fumehood (A1) would be not more than  $60 \text{ MBq}$ .

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# Distribution of radionuclides in the sediment of the southern Mediterranean Sea

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Keywords: Sediment, Southern Mediterranean, <sup>210</sup>Pb, <sup>137</sup>Cs, <sup>239,240</sup>Pu.

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Sediment samples were collected from the southern Mediterranean Sea in the framework of the IAEA's Technical Co-operation project in 2004 with the aim to investigate the distribution of artificial radionuclides such as plutonium isotopes, <sup>241</sup>Am and <sup>137</sup>Cs in the marine environment (Lee *et al.*, 2006). The age dating method derived from excess <sup>210</sup>Pb was used to interpret the sedimentary processes with Organic C analysis in the sediment column.

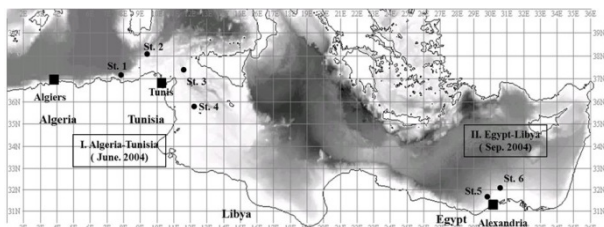


Figure 1. Sampling location in the southern Mediterranean Sea.

The sedimentation rates derived from excess <sup>210</sup>Pb in the southern Mediterranean Sea ranged from 2 to 3 mm yr<sup>-1</sup>. The most lowest sedimentation rate was found in the Sardinia Strait where is the most deepest sampling site.

Abrupt variations of sedimentation rate were observed in the Alexandrian (1.7-7.6 mm yr<sup>-1</sup>) and Nile River regions (2.0-7.2 mm yr<sup>-1</sup>), which might be induced by artificial activity such as Dam building in the river.

Organic C. content in the sediments ranged from 0.3 to 1.2 %. While the lowest Org. C contents have shown in the Sardinia Strait, the highest ones have found in the Nile River Region.

The activity ratios of <sup>238</sup>Pu/<sup>239,240</sup>Pu in the sediment have shown values close to global fallout ratios. The activity ratios of <sup>241</sup>Am/<sup>239,240</sup>Pu have shown the higher values than the global fallout. This is attributable to enhanced scavenging of <sup>241</sup>Am from the water column due to higher particle reactivity compared to

Pu. The southern Mediterranean has shown lower <sup>239,240</sup>Pu deposition density compared to the global fallout deposition. Especially, higher <sup>239,240</sup>Pu inventory appeared in the shelf near Nile River, which might be attributable to supply of <sup>239,240</sup>Pu bearing particles from Nile River. Most <sup>239,240</sup>Pu (18-97%) are deposited in sediment due to its enhanced scavenging from water column.

The <sup>241</sup>Am deposition densities are comparable to that of global fallout deposition. As <sup>239,240</sup>Pu shows, the higher <sup>241</sup>Am inventory appeared in the shelf near Nile River. More than 90% of <sup>241</sup>Am in the southern Mediterranean Sea are deposited in sediment due to its higher particle affinity.

<sup>137</sup>Cs deposition densities are much lower than that of global fallout deposition. This is related to shallow water depth because significant amounts of <sup>137</sup>Cs (> 85%) still retain in the water column (Lee *et al.*, 2006).

Table 1. Total inventory (seawater + sediment) of <sup>137</sup>Cs, <sup>241</sup>Am and <sup>239,240</sup>Pu in the southern Mediterranean Sea.

	St. 1	St. 2	St. 3	St. 4	St. 5	St. 6	Global fallout
<sup>137</sup> Cs* (Bq/m <sup>2</sup> )	1870	2200	1170	1170	600	810	2900
<sup>241</sup> Am (Bq/m <sup>2</sup> )	23	12	13	15	16	73	19
<sup>239,240</sup> Pu (Bq/m <sup>2</sup> )	48	32	16	15	19	167	47

<sup>137</sup>Cs\* has been decay-corrected (reference date April 2015)

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# Radiation dose assessment of $^{137}\text{Cs}$ in marine Fish of different habitats in the Aegean Sea - Greece

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Keywords: Cs-137 in seawater, Cs-137 in sediments, Cs-137 dose rates, Dose rates to Fish, Marine habitats  
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The 30-year retrospective synopsis of  $^{137}\text{Cs}$  activity concentrations in abiotic components from the Aegean Sea is used to assess the dose rates external and internal to seven representative fish species according to their habitats. Therefore, different exposure spaces as: a) infinitive for the sea water and sediment layers and b) semi-infinitive for the seawater-sediment interface are contemplated for the external dose rate estimations to natural marine biota. The concentration factors, calculated on the basis of real measurements, are used to estimate the internal dose rates in terms of three distinct habitats of Fish characterized as: pelagic, demersal and demersal-pelagic. For the dose rate calculations in seawater, different layers from surface to 1000 m have been differentiated, whereas the coastal to 250 m area has been used for the dose rate calculations from sediments. Besides, the time evolution of  $^{137}\text{Cs}$  activity concentrations from 1984 to 2015 in seawater and sediments are reported in terms of cold and warm period, with the radiological impact to marine biota selected from the three habitats to be estimated before and after the Chernobyl NPP accident.

Habitat	Species	Depth (m)	C.F. XE2	Dose rate XE-5
Pelagic	B. boops	to 350	5.77	67
Pelagic	S. flexuosa	to 130	0.73	85
Pelagic	S. pilchardus	to 180	6.54	76
Pelagic	T. trachurus	100-200 to 600	1.12	13
Demersal-Pelagic	P. erythrinus	20-420	2.3	18
Demersal-Pelagic	M. barbatus	10-420	0.65	5
Demersal	A. laterna	200-400	1.08	9

Table 1. Fish habitats and internal dose rates in various species due to  $^{137}\text{Cs}$  in seawater ( $\mu\text{Gy d}^{-1}$ )

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Station	Before Chernobyl	After Chernobyl
Saronikos gulf	$1.9 \cdot 10^{-5}$	$6.8 \cdot 10^{-5}$
Korinthiakos gulf	$1.9 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$
Evoikos gulf	$2.1 \cdot 10^{-5}$	$5.8 \cdot 10^{-5}$
Pagositikos gulf	$2.4 \cdot 10^{-5}$	$5.6 \cdot 10^{-5}$
Thermaikos gulf	$2.4 \cdot 10^{-5}$	$5.7 \cdot 10^{-5}$
Limnos	$2.4 \cdot 10^{-5}$	$12.1 \cdot 10^{-5}$
Lesvos	$2.2 \cdot 10^{-5}$	$9.5 \cdot 10^{-5}$
Milos	$1.9 \cdot 10^{-5}$	-----
Rhodes	$1.9 \cdot 10^{-5}$	$4.5 \cdot 10^{-5}$
Crete	$1.9 \cdot 10^{-5}$	$5.4 \cdot 10^{-5}$
North Aegean	$4.9 \cdot 10^{-5}$	$54.6 \cdot 10^{-5}$

Table 2. External dose rates due to  $^{137}\text{Cs}$  radiation in surface seawater in the Aegean Sea ( $\mu\text{Gy d}^{-1}$ )

Station	Before Chernobyl	After Chernobyl
Saronikos gulf	$1.0 \cdot 10^{-5}$	$4.2 \cdot 10^{-5}$
Korinthiakos gulf	$1.0 \cdot 10^{-5}$	$0.8 \cdot 10^{-5}$
Evoikos gulf	$3.5 \cdot 10^{-5}$	$9.3 \cdot 10^{-5}$
Pagositikos gulf	$3.5 \cdot 10^{-5}$	$9.3 \cdot 10^{-5}$
Thermaikos gulf	$0.2 \cdot 10^{-5}$	$14.5 \cdot 10^{-5}$
Limnos	$0.2 \cdot 10^{-5}$	$14.5 \cdot 10^{-5}$
Lesvos	$0.2 \cdot 10^{-5}$	$14.5 \cdot 10^{-5}$
Milos	$2.5 \cdot 10^{-5}$	-----
Rhodes	$2.5 \cdot 10^{-5}$	$4.2 \cdot 10^{-5}$
Crete	$1.0 \cdot 10^{-5}$	$4.2 \cdot 10^{-5}$
North Aegean	$4.8 \cdot 10^{-5}$	$15.1 \cdot 10^{-5}$

Table 3. External dose rates due to  $^{137}\text{Cs}$  radiation in surface sediments in the Aegean Sea ( $\mu\text{Gy d}^{-1}$ )

Station	Before Chernobyl	After Chernobyl
Saronikos gulf	$0.9 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
Korinthiakos gulf	$0.9 \cdot 10^{-5}$	$2.1 \cdot 10^{-5}$
Evoikos gulf	$2.1 \cdot 10^{-5}$	$7.6 \cdot 10^{-5}$
Pagositikos gulf	$2.1 \cdot 10^{-5}$	$7.5 \cdot 10^{-5}$
Thermaikos gulf	$0.5 \cdot 10^{-5}$	$10.1 \cdot 10^{-5}$
Limnos	$0.5 \cdot 10^{-5}$	$13.3 \cdot 10^{-5}$
Lesvos	$0.5 \cdot 10^{-5}$	$12.0 \cdot 10^{-5}$
Milos	$1.3 \cdot 10^{-5}$	-----
Rhodes	$1.6 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$
Crete	$0.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
North Aegean	$4.1 \cdot 10^{-5}$	$34.5 \cdot 10^{-5}$

Table 4. External dose rates due to  $^{137}\text{Cs}$  radiation in sediment - seawater intermediate phase in the Aegean Sea ( $\mu\text{Gy d}^{-1}$ )

# Seasonal and Spatial Distributions and Fluxes of $^{137}\text{Cs}$ , $^{226}\text{Ra}$ , $^{228}\text{Ra}$ , and $^{40}\text{K}$ in the Golden Horn Estuary, Istanbul

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Keywords: Golden Horn, sediment, sediment trap, radionuclide flux,  $^{137}\text{Cs}$ .

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Estuaries are important ecosystem because of being a transition zone between river environments and marine environments, high biodiversity and primer productivity. Considerably higher particulate matter flux is known to be one of the major factors effect radionuclide distributions in the estuaries. In the present study, radionuclide monitoring of surface sediment and suspended matter were implemented in the Golden Horn estuary. Surface sediment samples were collected at 14 locations in the Golden Horn Estuary during one year (Fig 1). The activity ranges of  $^{137}\text{Cs}$  in  $<63\ \mu\text{m}$  and  $>63\ \mu\text{m}$  fractions of sediment were found to be  $4.5\text{--}60.2\ \text{Bq kg}^{-1}$  and  $2.7\text{--}43.4\ \text{Bq kg}^{-1}$ , respectively (Fig 2-3). The activity ranges of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  in  $<63\ \mu\text{m}$  and  $>63\ \mu\text{m}$  sediment fractions were found to be  $11.5\text{--}31.4$ ,  $9.9\text{--}26.8\ \text{Bq kg}^{-1}$ ,  $15.4\text{--}35.5$ ,  $10.1\text{--}28.4\ \text{Bq kg}^{-1}$ ,  $227\text{--}824$ ,  $260\text{--}628\ \text{Bq kg}^{-1}$ , respectively. It is seen that the mean and the range of  $^{137}\text{Cs}$  activity concentrations for particle fractions ( $<63\ \mu\text{m}$  and  $>63\ \mu\text{m}$ ) are consistent with the levels of previous studies which performed in Bosphorus and Golden Horn (Kılıç and Çotuk, 2011; Kılıç et al., 2014).

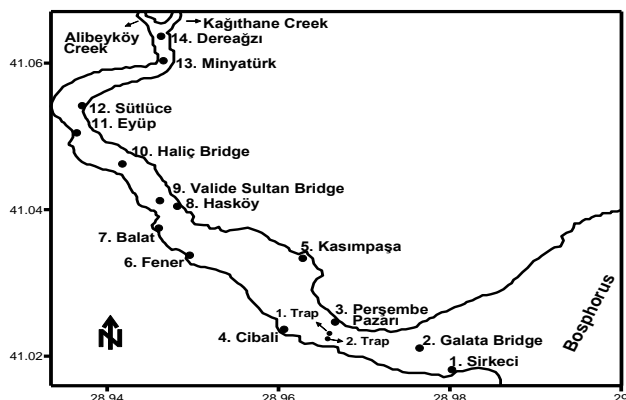


Figure 1. Sampling area and locations

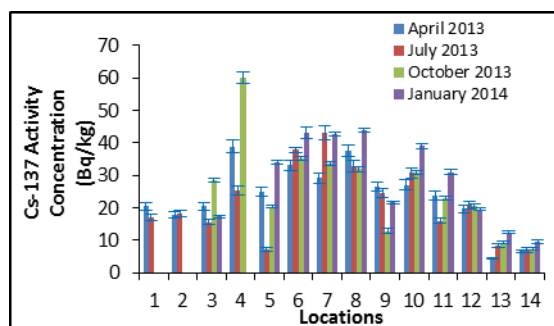


Figure 2.  $^{137}\text{Cs}$  activity concentration in  $<63\ \mu\text{m}$  fraction of sediment ( $\text{Bq kg}^{-1}$ , dry weight)

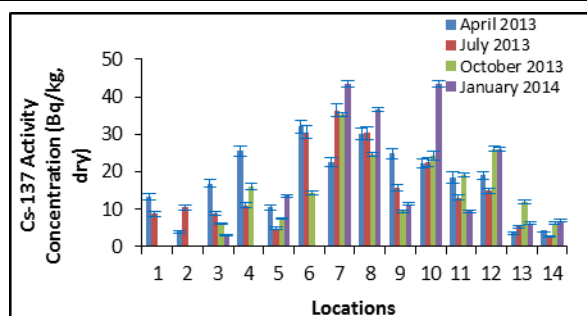


Figure 3.  $^{137}\text{Cs}$  activity concentration in  $>63\ \mu\text{m}$  fraction of sediment ( $\text{Bq kg}^{-1}$ , dry weight)

Two sediment trap samplers were established in Golden Horn in order to determine the fluxes of particulate matter and radionuclides. Flux rates of  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and sedimentation rate were seasonally determined through in 2013-14. Average particulate matter fluxes were measured to be  $62.2$ ,  $58.1$ ,  $70.9$ , and  $16.4\ \text{g/m}^2/\text{day}$  for Trap 1 in fall, winter, spring and, summer; respectively. Also similar measurements were performed for Trap 2. The results were found to be  $57.4$ ,  $66.0$ ,  $36.1$ , and  $12.8\ \text{g/m}^2/\text{day}$  in fall, winter, spring and, summer; respectively. The flux rates of  $^{137}\text{Cs}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  in sediment samples were also measured (Fig 4). The results indicated that sedimentation fluxes were lower than found in literature (Ergül and Topcuoglu, 2009).

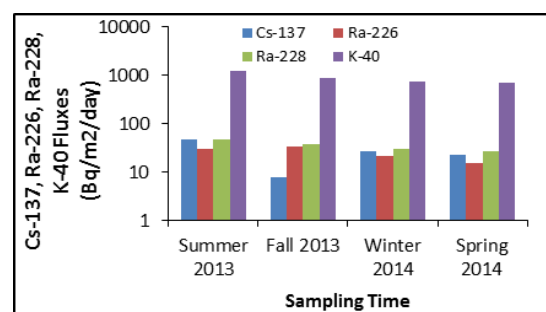


Figure 4. Radionuclide fluxes

## Acknowledgments

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# Medical I-131 in German Rivers

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## Introduction

<sup>131</sup>I is routinely used in nuclear medicine for the therapy of benign and malign thyroid diseases. Patients receive up to several GBq, taking advantage of the fact that the metabolism concentrates most of it in the thyroid, where the therapy delivers doses of several 100 Gy. In many countries, including Germany, patients are hospitalized during radioiodine therapy, and their excrements are collected in separate septic tanks where they are kept for decay before release to the sewer.

However, patients carry considerable amounts of <sup>131</sup>I to their homes after release from the hospital (in Germany, the maximum allowed activity is 250 MBq), and excrete part of this activity into the sewer. As a consequence, <sup>131</sup>I is routinely detected in wastewater treatment plants (WWTP) and their effluents to surface waters and can be found in river sediments near WWTP discharge points (Carvalho *et al.* 2013, Fischer *et al.* 2009, Rose *et al.* 2013).

In order to determine the total amount of <sup>131</sup>I transported to the North Sea, a combined experimental and modelling study was conducted, covering four main German rivers.

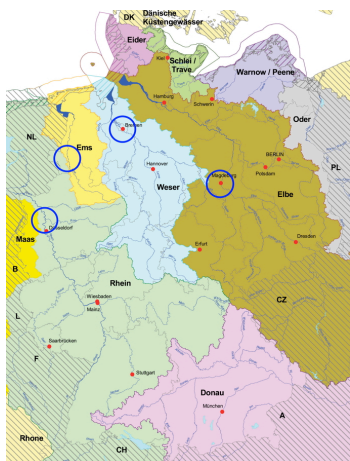


Figure 1. Rivers covered in the study, catchment areas and sampling points at sewage plants (graph: UBA).

## Experimental

The experimental part of the study involved monthly sampling of WWTP effluents and river water at selected sites of the rivers Rhine, Ems, Weser and Elbe over one year. Due to the low <sup>131</sup>I concentration in river water (in the mBq/l range), a precipitation technique from

50 l samples had to be used (Souti *et al.* 2014), whilst WWTP effluent could be measured directly by gamma spectrometry.

## Results

Results for WWTP effluent agreed with previous studies (Fischer *et al.* 2009). Considering population density in the catchment areas, frequency of radioiodine therapy and WWTP discharge rates, a mean discharge rate of 13 kBq per year and inhabitant could be calculated. This agrees fairly well with an earlier estimate of 40 kBq/(inh. yr) for input into the sewer system (Eschner *et al.* 2004), taking into account a measured reduction of <sup>131</sup>I concentration by 50% between WWTP in- and outflow (Fischer *et al.* 2009).

<sup>131</sup>I concentrations in river water are lower by several orders of magnitude and fall below the values expected from pure volumetric dilution of WWTP effluents with river water. The responsible effects could not yet be identified.

Yearly river loads can be calculated from water concentrations and river discharges. They sum up to ca. 250 - 270 GBq (see Table 1). This load is reaching the river estuaries, but the load reaching the North Sea is much lower due to radioactive decay.

Table 1. Measured WWTP effluent and river water <sup>131</sup>I concentrations, and total yearly river loads.

	Duisburg (Rhine)	Lingen (Ems)	Bremen (Weser)	Magdeburg (Elbe)
Effluent [mBq/l]	91 (<16–255)	117 (<14–503)	196 (84–419)	137 (39–279)
River water [mBq/l]	1.6 – 2.7	<0.7 – 5.9	<0.4–4.4	<0.5 – 2.5
Total load [GBq/yr]	234	4,0	10.8 – 12.2	2.7 – 17.3

## Acknowledgements

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# Chemical and isotope composition in selected Belarus mineral and therapeutic waters

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Keywords: Belarus mineral water, chemical composition, natural radionuclides, annual dose rates

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Belarus is a landlocked country in Eastern Europe covering 207 560 km<sup>2</sup> and its population amounts to about 10 million. From the geological point of view, the country consists of the crystalline fundament composed of igneous and metamorphic rocks (Archean-Proterozoic). The fundament morphology is very undulated. Generally, it occurs on the deep level but at some places it outcrops on the earth surface. The fundament is covered by the Paleozoic-Mezozoic sedimentary formations from a hundred to a few thousands meters of thick. At some places in the sedimentary formations there are intrusive dykes or batholiths. On the surface of the country there are Quaternary glacial sand, gravel and muds (Machnach, 2001). Such geological structure can form waters of different chemical and isotope compositions (Kudelski, 1994). In Belarus there are many centers using ground waters for the therapeutic purpose.

In this work, the waters from 22 therapeutic centers were investigated, at some centers there are two boreholes, the first exploits water from a shallow reservoir for consuming and the second water is taken from a deeper formation for bathing. In total, there were 22 drinking waters and 12 bath therapeutic ones, for every water the borehole depth, temperature, pH, Eh and conductivity of the water were measured and 5 liters of water samples were collected for chemical and isotopes analysis. For several waters the radon concentrations were also measured.

The depth of the water intakes varies between 100 to above 850 m under the earth surface, the pH ranges from 6.5 to 9.2 and the temperature of the waters from 9 to 15 °C, the conductivity and redox potential vary from 0.2 to above 187 µS/cm and

from -283 to 259 mV respectively. The waters in question belong to different hydrochemical types, but most of them are classified as the chloride-sodium (Cl-Na or Cl-SO<sub>4</sub>-Na). The mineralization (TDS) ranges from near 0.3 g/L up to 145 g/L. The uranium concentration is low and ranges from 0.5 mBq/L to 75 mBq/L for <sup>238</sup>U and 0.5 mBq/L to 289 mBq/L for <sup>234</sup>U. At several waters the activity of <sup>234</sup>U is significantly higher than that of <sup>238</sup>U ones. This is connected not only with the alpha recoil but also with the redox condition where Eh is below than -50 mV. The radium isotopes concentration ranges from below the limit detection (10 mBq/L) to ~ 4500 mBq/L for <sup>228</sup>Ra and from 5 mBq/L to 17600 mBq/L for <sup>226</sup>Ra. At one centers in shallow water <sup>226</sup>Ra activity is lower than that of <sup>228</sup>Ra, but in the deeper water <sup>226</sup>Ra > <sup>228</sup>Ra and at the second centers the phenomena are inverse. This fact could be explained by the different relation between uranium and thorium contents in the water bearing formations. Generally, the radon concentration is low and ranges from a few Bq/L to several tens or so, but at one water intakes the radon concentration reaches above 110 Bq/L. For the drinking waters the annual dose rates resulted from consuming 2 liters per day for adult were estimated. The statistic values of the measured natural isotopes of the all waters and annual dose rates of drinking ones are summarized in the Table 1. The annual dose rate estimated for majority of drinking waters is lower than 0.1 mSv. Based on the results of stable isotopes and tritium analysis, most of mineral waters originate from precipitation. It is worth adding that the most therapeutic waters contain some specific components such as Fe, Br, I and Rn which are useful for therapeutic hospitalization and relaxation.

Table 1. The statistic values of the measured TDS, radionuclides and annual effective dose caused by intake of the investigated drinking waters

	TDS	Ra-226	Ra-228	U-238	U-234	Rn-222	Dose rate
	g/L	mBq/L	mBq/L	mBq/L	mBq/L	Bq/L	mSv/y
Min	0.2	0.50	20.0	0.50	0.50	0.5	0.01
Max	18.2	2700	4500	75.2	289	28.9	2.83
Median	3.9	57.5	60.0	5.8	11.9	13.1	0.04
Arithmetical average	5.3	188	332	9.80	63.4	16.9	0.21
Geometrical average	3.6	47.4	75.0	4.08	14.4	3.6	0.05

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# Insight on the geochronology and heavy metal concentration of the Iacob Lake, Danube Delta, Romania

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**Keywords:** sediment dating, geochronology, <sup>210</sup>Pb (<sup>210</sup>Po method), heavy metal concentration  
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The sedimentation rates in the Danube Delta can be influenced in three main ways: by the modification in the sedimentation rate in shortening of the channels (changing the water debits and modifying the transported sediment), by building barrages on the Danube and by the erosion processes in the catchment area of the Danube after the building the Iron Gates. A detailed study with high time resolution on the sedimentation pattern changes in the Danube Delta, taking into consideration the heavy metal concentration in age concordance, was not made until now.

The aim of this study is to offer insight into the physical parameters, geochronology and sedimentation rates of the Iacob Lake (Danube Delta), as well as to determine the heavy metal concentration of each sediment layer.

The Iacob Lake is situated in the first part of the maritime Danube Delta, south to the Sulina branch. The outer part of its 9.98 km<sup>2</sup> surface is covered by vegetation, being responsible for the organic material content of the sediments. A total of four sediment cores (having lengths of approx. 60 cm each) have been taken from this lake (two at each sampling campaign).

After sub-sampling the sediment cores into 1-3 cm layers, these were weighted and dried for 24 h at 75°C, in order to determine physical parameters such as porosity, water content and bulk density. LOI measurements were also carried out: samples were put in porcelain crucibles in a muffle furnace at 350°C and 750°C, in order to determine the organic matter and the inorganic carbon content of each sub-sample.

High resolution gamma spectrometric measurements were carried out using an ORTEC GMX HPGe detector (FWHM of 1.92 keV at 1.33 MeV and a 0.5 mm Be window). The 46 keV gamma energy was used for determining <sup>210</sup>Pb, while <sup>226</sup>Ra was determined by using the 294 keV, 351 keV and 609 keV gamma energies. The sediment dating using the <sup>210</sup>Pb radionuclide has been accomplished via its progeny, <sup>210</sup>Po.

The <sup>210</sup>Po sources were prepared by adding to the measured 0.5g sediment 0.3 ml 100 Bq/l <sup>209</sup>Po tracer for determination of the chemical yield. Samples were then put to acidic digestion using a series of acids (2x10 ml 65% HNO<sub>3</sub>, 2 x 10 ml 35% HCl, 10 ml 6N HCl and 10x3 ml 8:1 35% H<sub>2</sub>O<sub>2</sub>: 35% HCl), after which they were deposited on high nickel content stainless steel discs (3 h at 85°C in a drying oven), interferences (iron ions) being eliminated by ascorbic acid. The obtained alpha sources were measured by an ORTEC SOLOIST 900mm2 PIPS detector, having a resolution of 19 keV and an ASPEC-

92 data acquisition system. The correctitude of the <sup>210</sup>Pb dating method was examined by an alternative dating procedure, namely by measuring <sup>137</sup>Cs at its 661 keV gamma energy line.

Heavy metal content of the sub-samples in one of the four sediment cores has been determined by atomic absorption spectrometry (AAS), the inspected heavy metals being Li, Mg, Al, K, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg and Pb.

Porosity and water content show an increasing, both having averages at 65%, and the dry and wet bulk densities show decreasing tendencies in concordance with sediment depth in case of the samples taken at the first sampling campaign, while the other two sediment cores show the contrary. The inorganic carbon content of the sediment cores is usually higher in the deeper layers, while organic carbon content shows fluctuations in concentration throughout the entire sediment core in all cases.

The measured <sup>226</sup>Ra concentration had an average of 19.4 Bq/kg, while the atmospheric <sup>210</sup>Pb content decreased from an average of 64.2 Bq/kg. The correctitude of the used method was confirmed by using the <sup>137</sup>Cs radionuclide, its two peaks (1963 – nuclear tests and 1986 – Chernobyl accident) matching the ages obtained by the CRS method.

In spite of the differences shown in the sedimentation patterns, all sediment cores show increased values for both linear and mass sedimentation for the 2005-2006 years and for the 1970-1980 period. Studies show, that there were two major floods between 2005-2006 and six between 1970-1980 (all of these have the maximum water discharges over 10000 m<sup>3</sup>/s).

Heavy metal concentrations were measured, average results are as follows: 16.933 ppm Li, 3.955 % Mg, 10.726 % Al, 2.275 % K, 28.554 ppm Cr, 291.408 ppm Mn, 14.09 % Fe, 6.006 ppm Co, 14.924 ppm Ni, 22.883 ppm Cu, 8.289 ppm As, 401.883 ppb Cd and 245.45 ppb Hg. In case of Zn and Pb, half of the measures samples were below the limit of detection, their highest value being 78.469 ppm and, respectively, 11.575 ppm. The obtained results are on average 3-4 times higher than the values measured in water during the 2005-2009 period by the Black Sea Scientific Network.

## Acknowledgments

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# Levels of natural radionuclides in Pit lakes from countries in Africa and Europe

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During exploitation by open-pit mining, the water table is suppressed to avoid the flooding of active mines. However, when mining activity ceases, the water table recovers its original position, flooding the open pits and giving rise to mine pit lakes. The environmental problem arise because these waters can be affected by Acidic Mine Drainage (AMD), having high/very high concentration of heavy metals in solution. This is a well known problem worldwide but hardly nothing has been investigated regarding natural enhancement in <sup>238</sup>U and <sup>232</sup>Th isotopes in these water bodies (apart from Uranium mining sites). This work pretends to cover this gap. One study in Pit leaks along the Iberic Piritic Belt in the south-west of Spain, was carried out by our research group [Manjón et al 2013]. They found activity concentrations of <sup>238</sup>U that ranged from 1.1-1110 mBq/L, <sup>234</sup>U ranging from 39-1750 mBq/L in Pit lakes with very low pH values (2.21 to 2.71) associated mainly to iron sulphide minerals.

In this work, alpha spectrometry with PIPS detectors (in superficial water and sediment samples from shoreline), and gamma spectrometry with HpGe coaxial detector (in sediment samples) were performed. In addition, other parameters in water samples as pH, redox potential (Eh), Electric Conductivity (EC) or Total Dissolved Solids (TDS) were measured. The main aim was to carry out a first screening about levels in natural radionuclides in a set of pit lakes placed at the Upper Moulouya Mining District (Eastern Morocco) and also in the north of Europe (several mine districts in central and southern Sweden).

Table 1. Activity concentration (mBq/L) of U isotopes in water from Moroccan pit lakes.

Sample	<sup>238</sup> U	<sup>234</sup> U	Ph
HM4	1027±42	4526±185	9,15
HM5	443±8	1921±32	9,57
HM7	235±14	1267±62	9,23

Table 1 shows U isotopes in a set of pit lakes with similar values that the ones found in Spain. Environmental levels in water from non-affecting scenarios around the world is about 30-50 mBq/L for both U isotopes. The levels found here are between ten and hundred times higher. However for Th (<sup>232</sup>Th, <sup>230</sup>Th) or <sup>210</sup>Po isotopes, the activity concentration not only in waters but also in sediments, were found at environmental non-affected levels. It is worth to mention that pH ranged from 9.15-9.57, what implies a complete different U chemical behaviour at these water bodies compared to the Spanish situation. Alkaline pH values and elevated bicarbonate concentrations in oxidized ground and surface waters favour the stabilization and mobilization of uranium as uranyl-carbonate complex, [Abdelouas et al.1998] while Th will be scavenged from the water column. In contrast, the predominant species in acid, oxygenated waters are the uranyl ion and uranyl-sulfate complex [Wanty et al. 1999]. Thus acid or alkaline oxygenated mine waters may carry significant concentrations of dissolved uranium.

Additional results will be shown with the first set of results about pit lakes from central and southern Sweden and all the three countries in this environmental framework will be compared.

## Acknowledgments

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# Approximate dating of a sediment core obtained from Tuz Lake in Turkey using Cs-137 isotopic measurements

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Tuz (Salt) Lake is the second largest lake in Turkey with an area of 1600 km<sup>2</sup> surrounded by the cities Konya, Aksaray, and Ankara. Besides the lake hosts many life forms such as flamingos and many endemic species that are worldwide endangered, 70% of Turkey's salt demand is supplied from Tuz Lake.

Pollutants might be carried into Tuz Lake from industrial and urban sources by streams entering the lake. It is also possible that various pollutants might reach the lake through the drainage canal of General Directorate of State Hydraulic Works in Konya. Sediments accumulated on the lake bottom can give information both about the current status and the historical change of probable pollution.

Dating of the sediment layers is important to understand the possible causes of probable elemental concentration changes in the lake sediments. There are various techniques used in sediment dating studies. For example, <sup>210</sup>Pb isotope is the most often used one for accurate sediment age determination. However, this isotope requires quite complex chemical separation operations for accurate determination and is out of the scope of this study. Another useful technique is the use of reference horizons from the atmospheric atomic bomb tests and Chernobyl nuclear reactor accident through Cs-137 measurements. This radioactive isotope does not exist naturally and only released from nuclear operations. The atmospheric atomic bomb tests were performed between 1950 and 1965. In literature, the first observable signal of this isotope in sediment and soil samples is given as 1954 while the peak concentration years are given as 1963-1964. Another reference horizon is given as Chernobyl nuclear reactor accident in 1986 (Zhiyanski *et al.*, 2008).

A sediment core obtained from the dry lake bed in summer months was analyzed by dividing it into pieces one centimeter thick each. Each piece was separated into three groups according to particle sizes as fine particles smaller than 63 micrometers, coarse particles between 63-212 micrometers, and large particles greater than 212 micrometers.

In this study, a total of 15 samples down from the surface were counted in a gamma ray spectrometry system for their Cs-137 contents. These measurements were performed at the Kucukcekmece Nuclear Research Center of the Turkish Atomic Energy Authority located in Istanbul. The goal of these analyses was to obtain peaks in the above mentioned reference years. Three samples from each size fractionated group (<63 µm, 63-212 µm,

>212 µm) were pre-analyzed to determine the strongest Cs-137 counting rates among them and Group-B (63-212 µm) samples were seen to have the strongest gamma signal per unit mass. Therefore, this group of samples was used in further analyses. The gamma ray spectrometry system was calibrated by a multi-nuclide standard source.

As can be seen from the Figure 1, the first sample with detectable Cs-137 level was the sample between 8 and 9 cm. Therefore, this depth is considered to represent the year 1954. The first peak observed for the samples representing 5-6 cm and 6-7 cm is considered to represent the years 1963-1964. The second peak for the sample between 1 and 2 cm is considered to represent 1986.

Based on this graph with reference years 1954, 1963-1964, and 1986, the approximate sedimentation rate for the sediment core was calculated as 0.18 centimeters per year.

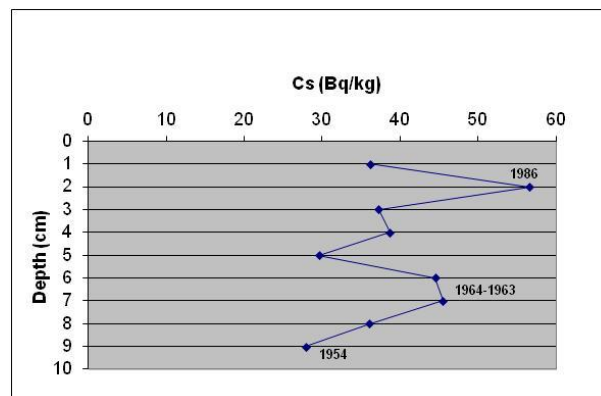


Figure 1. Decay Corrected <sup>137</sup>Cs concentrations vs Depth and reference years.

## Acknowledgments

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## Variability in the uranium isotopic ratios in the vicinity of former uranium mining and milling sites

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Uranium (U) mining activities in Mailuu Suu (Kyrgyzstan) started in 1946 and lasted until 1968. Large volumes of residues from the mining and milling of the minerals were produced and disposed in near-surface impoundments in the vicinity of the mines and mills. A typical problem arising from these tailings is the leaching of contaminants into surface and ground waters..

In order to assess the potential contamination of the water resources, a large number of samples were collected from the drinking water distribution system (DWDS), rivers, shallow aquifers and seepage/drainage water from the tailings. Soil and river sediment samples were also taken (Corcho Alvarado et al. 2014).

The samples were analysed by using a double focusing sector field Inductively Coupled Plasma Mass Spectrometry (sf-ICP-MS, Element2, Finnigan)..

We observed a large variation in  $^{234}\text{U}/^{238}\text{U}$  isotope ratios in the water samples, with values near equilibrium at the mine tailings and far from equilibrium outside this area. Large variability in the  $^{234}\text{U}/^{238}\text{U}$  isotopic ratio, in the percentage level, has long been observed in the environment. These variabilities are known to be caused by selective leaching of  $^{234}\text{U}$  atoms occupying radiation damaged crystalline lattice sites upon alpha decay of  $^{238}\text{U}$  (Suksi et al. 2006).

The higher U concentrations of 159 and 6820 µg/L were measured in the drainage water from the mine tailings. U isotopes were in their natural radioactive equilibrium (activity ratios of  $^{234}\text{U}/^{238}\text{U} = 1$ , and  $^{235}\text{U}/^{238}\text{U} = 0.047$ , Fig. 1). Concentrations of U of 3 to 10 µg/L were observed in four groundwater samples, with an excess of  $^{234}\text{U}$  in all them. The U content in the samples taken from the DWDS varied in a narrow range from 0.27 to 0.34 µg/L.  $^{234}\text{U}$  was enriched in all the samples. Upstream of the former mining areas and the tailings

(No. 21), the U content in river water does not differ much from the DWDS. However, downstream of the mine tailings, the U concentrations in river water were a magnitude higher with values up to 4.2 µg/L. An excess of  $^{234}\text{U}$  was observed in all the samples, except in the river water sampled downstream.

The  $^{235}\text{U}/^{238}\text{U}$  isotope ratios in the water samples depicted very small variations, in the permil level. Recent studies have shown that variability in the  $^{235}\text{U}/^{238}\text{U}$  ratio over a range of a few permil occur in the environment (Stirling et al. 2007; Weyer et al. 2008; Brennecka et al. 2010). This variability of the  $^{235}\text{U}/^{238}\text{U}$  ratio may be related to the occurrence of low temperature redox reactions (Stirling et al. 2007; Weyer et al. 2008; Brennecka et al. 2010)

A large variation in U isotopic ratios in water was observed, with values near equilibrium at the mine tailings and far from equilibrium outside this area. These results highlight the potential use of the U isotope ratios as indicators of the origin of U contamination in Mailuu Suu.

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## Are drinking water guidelines for tritium adequate to protect aquatic biota?

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Tritium activity concentration guidelines in drinking water vary throughout the world, ranging from 100 Bq L<sup>-1</sup> as an action level for further assessment (European Union) to 76,103 Bq L<sup>-1</sup> (Australia). Currently, in Canada the tritium drinking water limit is 7,000 Bq L<sup>-1</sup>.

In order to evaluate whether the current Canadian drinking water limit is protective of aquatic biota, we examined the effect of *in vivo* exposure to 10-100,000 Bq L<sup>-1</sup> tritiated water in trout and fathead minnows in lab and field settings, respectively. Further *in vitro* studies were conducted to assess the effects of this exposure regime on cell viability, cell growth, DNA breaks,  $\gamma$ -H2AX expression and micronucleus frequency in channel catfish peripheral B-lymphoblast and fathead minnow testis cell cultures.

Exposure to 10 - 100,000 Bq L<sup>-1</sup> of tritium consistently caused no detrimental effects on fish growth, fish condition, cell viability and cell growth. Different types of dose-responses were observed

when molecular markers were assessed. These studies showed that DNA strand breaks evaluated using the comet assay, tended to increase with the level of exposure while gamma-H2AX expression was only slightly elevated in exposed groups compared to controls regardless of the level of exposure. The micronucleus frequency and membrane fatty acid composition data provided evidence for adaptive responses.

The data collected to date demonstrate that, within cells, some molecular processes are activated in response to low levels of tritium exposure. However, there was no evidence of detrimental effects on cells and on fish. In combination our results suggest that dose-responses to tritium at low concentrations follow a threshold as opposed to a linear pattern in live fish, and that the current Canadian drinking water limit, established for human protection, seems adequate to ensure the protection of aquatic biota.

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## Radionuclide concentrations in mussels (*Mytilus galloprovincialis*) collected from the eastern black sea region of Turkey in 2014

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Many anthropogenic radionuclides entered into the Black Sea after Chernobyl nuclear power plant accident. The accident released a large activity of <sup>137</sup>Cs to the atmosphere. The radioactivity levels in soft tissues of mussels (*Mytilus galloprovincialis*) collected during the period of February - November 2014 seasonally from twelve different stations within the border of the Eastern Black Sea Region of Turkey were determined using a gamma spectrometer equipped with HPGe detector. The

activity concentrations obtained for <sup>226</sup>Ra, <sup>232</sup>Th, <sup>137</sup>Cs and <sup>40</sup>K are given in the unit of Bq/kg. The results have been compared with the previous results.

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# Fukushima Accident and Environmental Radioactivity

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*Keywords: Fukushima accident, radionuclide emission,  $^{137}\text{Cs}$  deposition,  $^{137}\text{Cs}$  activity in air.*  
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After the 2011 Great East Japan Earthquake and resulting tsunami, severe accident occurred at the Fukushima Dai-ichi nuclear power plant (FDNPP), which caused atmospheric emission and ocean discharge of huge amounts of radionuclides (Povinec et al., 2013). In order to assess environmental effects of FDNPP-derived radionuclides, Japanese Government, Local Governments, Research Institutes and Universities have carried out emergency environmental monitoring of the FDNPP-derived radionuclides using monitoring networks and studied environmental behaviors of the FDNPP-derived radionuclides. As a result, we have better understanding about total emission amounts of radionuclides from the FDNPP accident, including atmospheric emission and direct discharge of contaminated water, and environmental effects such as transport processes of radionuclides in atmosphere, ocean and land, spread of contaminated area, and fates of radionuclides in future.

In this paper, we review results of the environmental monitoring and environmental radioactivity studies in Japan. Although environmental radioactivity studies include all of natural processes in atmosphere, land and ocean in addition to human activities such as decontamination, we present here mainly the atmospheric processes of the FDNPP-derived radionuclides in aspects of environmental monitoring.

After the FDNPP accident, extremely high activities of anthropogenic radionuclides were observed in surface air near the sites, and radioactive cloud spread out in northeast region of Honshu Island, Japan. In late March and early April 2011, the FDNPP-derived radionuclides were detected in surface air at many sites in North America, Europe and Asia. These observations suggest that the radioactive cloud originating from the FDNPP spread out in the world. Major radionuclides released from the FDNPP were radioxenon, radioiodine and radiocesium. The atmospheric effects of radiocesium have still continued now at the monitoring sites within about 200 km from the FDNPP.

The radionuclides emitted into atmosphere were deposited on land surface by wet and dry deposition processes. The highly contaminated area, which was realized by aerial monitoring and radioactivity contamination mapping program, extended to more than 30 km northwest from the FDNPP. The radioactivity-contaminated areas were affected by land topography.

Japanese government has carried out continuous monitoring of deposition (rainwater and falling dust) of radionuclides using a monitoring network. A typical time sequence of the monthly  $^{137}\text{Cs}$  deposition is as follows: 1. Marked high  $^{137}\text{Cs}$  deposition ( $3.34 \text{ MBq m}^{-2}$ ) occurred in March 2011 at the Futaba monitoring site, where is located about 5 km of the FDNPP. 2. The monthly  $^{137}\text{Cs}$  deposition at Futaba rapidly decreased from March to June 2011 as a result of cease of major radioactivity emission from the FDNPP. 3. The decrease rates of  $^{137}\text{Cs}$  deposition slowed down from July to October 2011 and a minimum monthly  $^{137}\text{Cs}$  deposition ( $1.55 \text{ kBq m}^{-2}$ ) occurred in October 2011. 4. After then monthly  $^{137}\text{Cs}$  deposition gradually increased to a maximum ( $19.5 \text{ kBq m}^{-2}$ ) in February 2012. 5. After a peak in February 2012, the monthly  $^{137}\text{Cs}$  deposition deceased and reached a minimum in August 2012. The monthly  $^{137}\text{Cs}$  deposition increased from September and reached a maximum in winter, 2013 (January 2013:  $18.9 \text{ kBq m}^{-2}$ ). 6. The similar seasonal variation of the monthly  $^{137}\text{Cs}$  deposition occurred in 2013. 7. The annual  $^{137}\text{Cs}$  deposition decreased from 2011 to 2014. The FDNPP-derived  $^{137}\text{Cs}$ , however, has been still detected in deposition samples in January 2015 at most of the monitoring stations within 300 km of the FDNPP.

A cause of higher  $^{137}\text{Cs}$  deposition and higher surface  $^{137}\text{Cs}$  concentrations in current air was presumed to be resuspension of  $^{137}\text{Cs}$ -enriched particles (Hirose, 2013). However, in addition to monitoring results, chemical speciation of  $^{137}\text{Cs}$  bearing particles in airborne particulate matter and soil suggests that continuous uncontrolled emission from the FDNPP and other processes such as biomass burning have to be considered as additional potential sources controlling current higher  $^{137}\text{Cs}$  deposition and surface activities.

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# <sup>134</sup>Cs and <sup>137</sup>Cs in the North Pacific Ocean derived from the TEPCO Fukushima Dai-ichi Nuclear Power Plant accident in March 2011

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**Keywords:** Radiocaesium, North Pacific Ocean, accident, transport processes

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On 11 March 2011, an extraordinary earthquake of magnitude 9.0 centered about 130 km off the Pacific coast of Japan's main island of Honshu, at 38.3°N, 142.4°E, was followed by a huge tsunami with waves reaching up to 40 m height in Iwate region and about 10 m in Fukushima region. One of the consequences was a station blackout (total loss of AC electric power) at the Fukushima Dai-ichi Nuclear Power Plant (FNPP1). The station blackout developed into a disaster that left three of the six FNPP1 reactors heavily damaged and caused radionuclides to be discharged into the air and ocean (Tsumune, Tsubono et al. 2013).

We collected 2 - 10 litre surface seawater samples at more than 300 stations in the North Pacific Ocean and two coastal stations at Tomioka in Fukushima Prefecture and Hasaki in Ibaraki Prefecture. Water column samples were also collected at 24 stations. An improved ammonium phosphomolybdate, AMP, procedure developed by one of the authors was used to extract radiocaesium from the samples. With this improvement of AMP procedure, the weight yield of AMP/Cs compound generally exceed 99 % for 2 litre samples as well as radiochemical yield of radiocaesium. The activities of AMP/Cs compound were measured at the Ogoya Underground Facility of the Low Level Radioactivity Laboratory of Kanazawa University using high-efficiency, well-type ultra low background Ge-detectors.

After July 2012, the activities of <sup>137</sup>Cs in surface water at near FNPP1 site were still kept around 1000 Bq m<sup>-3</sup> which corresponds about 10 GBq day<sup>-1</sup>. At Hasaki which locates 180 km south of FNPP1, two sudden increases of radiocaesium were observed in 2011 and 2012 due to rapid southward transport of FNPP1 derived radiocaesium due to disappearance of warm water eddy at around end of May in both year. In 2014 at Hasaki, <sup>137</sup>Cs activity remains at a few Bq m<sup>-3</sup> which might be affected not only ongoing FNPP1 flux of radiocaesium but widely existed slightly contaminated surface waters including south of Kuroshio (Figure 1).

A zonal speed of FNPP1 derived radiocaesium in surface water at mid latitude in the North Pacific Ocean was 7 km day<sup>-1</sup>, 8 cm s<sup>-1</sup> until March 2012 (Aoyama, Uematsu et al. 2013), however it after March 2012 till August 2014 was ca. 3 km day<sup>-1</sup>, 3.5 cm s<sup>-1</sup>. Although observed decrease of zonal speed of Fukushima derived radiocaesium in surface water is larger rather than the decrease of zonal current speed in the eastern North Pacific Ocean, the latter explains the tendency for zonal

speed of Fukushima derived radiocaesium to decrease (Figure 2). In terms of inventory, about 5 % of released radiocaesium transported in the surface layer and arrived at west coast of US continent, while 95 % of them were already transported in the ocean interior.

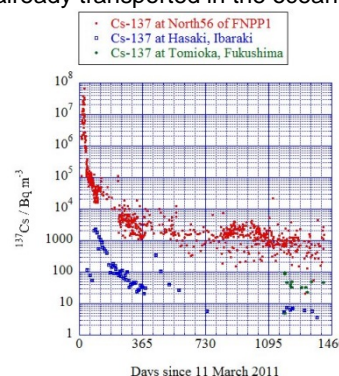


Figure 1. <sup>137</sup>Cs activity in surface seawater at North56 point at FNPP1 observed by TEPCO, and those at Tomioka and Hasaki.

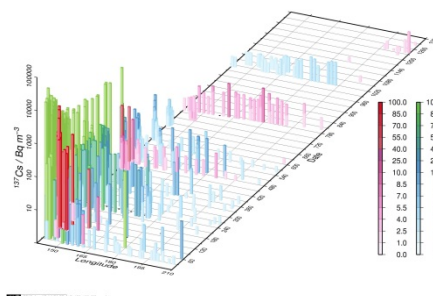


Figure 2. <sup>137</sup>Cs activity in surface seawater in the North Pacific Ocean during the period from 11 March 2011 to the end of 2014.

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# Applications of Radon as a Tracer in Atmospheric Studies: Lessons Learned and Future Challenges

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Radon-222 and its progenies have provided a wealth of information on the sources of air masses and aerosols, residence times and removal rate constants of aerosols and stability and vertical movements of air masses in the planetary boundary layer. The utility of  $^{222}\text{Rn}$  as an atmospheric tracer is due to the fact that its mean-life (5.51 d) is long compared to typical turbulent time scales ( $< 1$  hr), but short enough to constrain  $^{222}\text{Rn}$  concentrations in the free troposphere. Its mean life is generally comparable to the transit time of air masses across major continents and/or ocean but much shorter compared to the mixing time scale of the atmosphere and hence, it is widely dispersed in the atmosphere.

The effective utility of radon as an atmospheric tracer critically depends on our understanding of the radon emanation rate from earth's surface. However, the radon emanation estimates from soils and ocean surface are highly variable. The radon emanation rates from

continents vary over an order of magnitude, with the lowest values from the polar region and highest values from select regions in Northeast Asia (mainly Japan). The radon gradient in the atmosphere, from planetary boundary layer to the lower stratosphere, observed at different latitudes and longitudes will be discussed. Although considerable efforts been made in predicting the seasonal distribution of radon in the atmosphere, there are still an order of magnitude difference in the prediction among a group comprised of 11 different modelers and thus the present models are not well constrained. Occasionally very high concentrations of  $^{222}\text{Rn}$  have been observed both in the Arctic and Antarctic regions (known as 'radonic storms'), but the cause for this remains unknown. In this presentation, a possible mechanism for the presence of very high  $^{222}\text{Rn}$  concentrations and possible will be presented to explain the radonic storms.

# Four-years, regionalscale simulation of $^{137}\text{Cs}$ radioactivity in the ocean following the Fukushima Daiichi Nuclear Power Plant accident

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A series of accidents at the Fukushima Dai-ichi Nuclear Power Plant (1F NPP) following the earthquake and tsunami of 11 March 2011 resulted in the release of radioactive materials to the ocean by two major pathways, direct release from the accident site and atmospheric deposition.

We reconstructed spatiotemporal variability of  $^{137}\text{Cs}$  activity in the ocean for four years by the comparison model simulations and observed data. We employed a regional scale and the North Pacific scale oceanic dispersion models, an atmospheric transport model, a sediment transport model, a dynamic biological compartment model for marine biota and river runoff model to investigate the oceanic contamination.

Direct releases of  $^{137}\text{Cs}$  were estimated for four years after the accident by comparing simulated results and observed activities very close to the site. The estimated total amounts of directly released was  $3.6 \pm 0.7$  PBq. Directly release rate of  $^{137}\text{Cs}$  decreased exponentially with time by the end of December 2012 and then, was almost constant. Decrease rate were quite small after 2013. The daily release rate of  $^{137}\text{Cs}$  was estimated to be the order of magnitude of  $10^{10}$  Bq/day by the end of March 2015 (Table 1). The activity of directly released  $^{137}\text{Cs}$  was detectable only in the coastal zone after December 2012. Simulated  $^{137}\text{Cs}$  activities attributable to direct release were in good agreement with observed activities (Figure 1), a result that implies the estimated direct release rate was reasonable. Annual averaged distribution of simulated  $^{137}\text{Cs}$  activity in 2013 is good agreement with the one in 2014. Annual change of dispersion process off Fukushima coast is small. And seasonal changes are similar both in 2013 and 2014. Stratification in summer affects dispersion process in the simulation. We can predict the distribution of radionuclides released from 1F NPP in the future.

There is no observed data of  $^{137}\text{Cs}$  activity in the ocean from 11 to 21 March 2011. Observed data of marine biota should reflect the history of  $^{137}\text{Cs}$  activity in this early period. We reconstructed the history of  $^{137}\text{Cs}$  activity in this early period by considering atmospheric deposition, river input, rain water runoff from the 1F NPP site and absorption in sediment. The comparisons between simulated  $^{137}\text{Cs}$  activity of marine biota by a dynamic biological compartment and observed data also suggest that simulated  $^{137}\text{Cs}$  activity attributable to

atmospheric deposition was underestimated in this early period. In addition, river runoff model simulations suggest that the river flux of  $^{137}\text{Cs}$  to the ocean was effective to the  $^{137}\text{Cs}$  activity in the ocean in this early period. The sediment transport model simulations suggests that the inventory of  $^{137}\text{Cs}$  in sediment was less than 10% of total released  $^{137}\text{Cs}$ . Sediment is not dominant sink of  $^{137}\text{Cs}$  in the ocean.

Table 1. Estimated release rate.

Term	Averaged $^{137}\text{Cs}$ activity (Bq/m <sup>3</sup> ) adjacent to 1F NPP	Release rate (Bq/day)
2011/3/2-6-4/6	$1.1 \times 10^7$	$2.0 \times 10^{14}$
2013/1/1-12/31	$1.0 \times 10^3$	$2.0 \times 10^{10}$
2014/1/1-12/31	$5.8 \times 10^2$	$1.2 \times 10^{10}$
2015/1/1-3/31	$3.0 \times 10^2$	$6.0 \times 10^9$

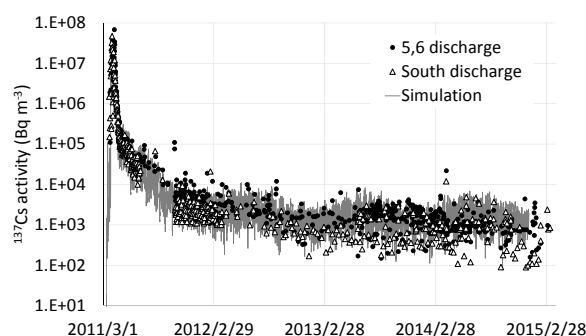


Figure 1.  $^{137}\text{Cs}$  activity adjacent to 1F NPP.

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# <sup>137</sup>Cs and tritium concentrations in seawater off the Fukushima prefecture: results from the SOSO 5 rivers cruise (October 2014)

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Keywords: Fukushima, caesium, tritium, seawater, estuaries

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, which occurred in northeastern Japan on March 11, 2011, resulted in the releases of large amount of various radionuclides especially cesium isotopes (<sup>137</sup>Cs half-life 30.2y and <sup>134</sup>Cs half-life 2.06y) both in the atmosphere and in the oceans. Various authors have observed that caesium have been transported from contaminated watershed to rivers (e.g. Nagao et al., 2013). Three and a half years after the accident this study has been conducted in order to investigate the river influence on coastal areas.

In October 2014, the cruise SOSO 5 Rivers took place off the coast of the Fukushima prefecture. The sampling was targeted off the mouths of 5 coastal rivers located North of the FDNPP and whose watershed were strongly contaminated by the FDNPP accident fallout i.e. Mano, Nitta, Ota, Odaka and Ukedo rivers.

Seawater was sampled along radials comprising 5 stations in front of each of these 5 rivers. Seawater was sampled on surface and 1m above bottom. In addition one water sample was realised in 4 estuaries (the Ukedo estuary was not accessible at that time). All the samples were filtered on 0.45µm. An improved ammonium phosphomolybdate, AMP, procedure (Aoyama and Hirose, 2008) was used to extract radiocaesium from the samples. With this improvement of AMP procedure, the weight yield of AMP/Cs compound generally exceed 99 % for 2 litre samples as well as radiochemical yield of radiocaesium. The activities of AMP/Cs compound were measured at the Ogoya Underground Facility of the Low Level Radioactivity Laboratory of Kanazawa University using high-efficiency, well-type ultra low background Ge-detectors. One liter of seawater underwent tritium analysis through electrolytic enrichment at the tritium laboratory (Miami, USA).

Both <sup>134</sup>Cs and <sup>137</sup>Cs were detected in all the samples demonstrating a contamination from the FDNPP accident releases. Generally the concentrations were higher at coastal sites and decreased with distance from the coast, and were higher in the surface layer compared to the bottom layer. The relationship between <sup>137</sup>Cs and <sup>134</sup>Cs decay corrected to the date of the accident showed that pre-Fukushima <sup>137</sup>Cs activities due to global fallout from nuclear bomb testings were close to 1.5 Bq.m<sup>-3</sup> in agreement with Aoyama et al. (2008). The <sup>137</sup>Cs/<sup>134</sup>Cs activity ratio were close to 1, a value consistent with the ratio observed nearby the release point after the accident (Buesseler et al., 2011)

Regarding tritium only a part of all samples (11) were analysed (table 1). The tritium levels are quite low but higher levels always characterized estuarine waters.

In seawater they ranged from 1.48±0.09 in front of the Mano river to 0.57±0.09 TU at 6.7 nautic miles of the Nita mouth (Nitta 5). These results underlined the fact that out of rivers influence seawater tritium content are less than 1 TU which corresponds to background level in the marine environment. In rivers tritium contents ranged from 4.10±0.14 in the Ota estuary to 1.81±0.09 in the Odaka estuary. These levels are in agreement with what is known from continental freshwater, the continental tritium reservoir being higher than the marine one. In Japan, Matsumoto et al (2013) considered that levels up to 6TU are conservative estimate of the pre-Fukushima background level. Therefore our dataset does not show any influence of the tritium released by the Fukushima accident on these rivers or on coastal waters north FDNPP in October 2014.

Location	Date (yymmdd)	TU	eTU
Mano Estuary	14/10/11	3.28	0.11
Mano 1 surface	14/10/09	1.48	0.09
Nitta Estuary	14/10/11	3.89	0.13
Nitta 1 surface	14/10/19	0.89	0.09
Nitta 5 surface	14/10/19	0.57	0.09
Ota Estuary	14/10/11	4.10	0.14
Odaka Estuary	14/10/11	1.81	0.09
Odaka 1 surface	14/10/18	0.99	0.09
Odaka 5 surface	14/10/18	0.68	0.10
Ukedo 1 surface	14/10/17	0.92	0.09
Ukedo 5 surface	14/10/17	0.70	0.09

Table 1: Tritium content (±1 sigma) in rivers and coastal rivers (SoSo5 rivers project-October 2014).

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# Biokinetics of radiocesium depuration in marine fish inhabiting the vicinity of the Fukushima Dai-ichi Nuclear Power Plant

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The radiocesium were released to the coastal water by accident of the Fukushima Dai-ichi Nuclear Power Plant (1FNPP) on March 2011. The introduced radiocesium into seawater was transferred rapidly to marine fish inhabiting the coastal water vicinity of the 1FNPP, resulting the elevated radiocesium concentration even at the four years after from the accident. The radioactivity monitoring for fish inhabiting near the 1FNPP started only after two years later. Modelling approach was applied to reconstruct the radioactivity levels in the marine biota (Vives i Batlle et al., 2014), however the analysis with food chain transfer was not included in the evaluation. For the better understanding of radiocesium biokinetics in marine biota, the reconstruction of radioactivity levels including food chain transfer is necessary.

In this study, dynamic biological compartment model of food chain transfer (Tateda et al., 2013) was applied to simulate the radiocesium levels in fish inhabiting the vicinity of the 1FNPP to reconstruct <sup>137</sup>Cs concentration from the beginning of the accident, for understanding of the radiocesium biokinetics in fish, especially the determining factor of the maximum level and rapidity of the depuration.

For the calculation of fish inhabiting inside the port of 1FNPP, the reported <sup>137</sup>Cs concentrations in seawater collected from the port by TEPCO monitoring were used. For the evaluation of the radiocesium levels in fish that swam into the port from outside, the Regional Ocean Model System ROMS was used to simulate <sup>137</sup>Cs concentrations in seawater at TEPCO monitoring point T-E1 1km off 1FNPP. The sources used were direct leaked radioactivity 3.6 PBq (Tsumune et al., 2013) and continuous release 50 GBq d<sup>-1</sup> after 1 November 2011 (Kanda, 2013). For the verification of the simulated values, monitored seawater concentrations at T-E1 were used. The <sup>137</sup>Cs concentrations in fish inhabiting inside the port was simulated by observed seawater concentration, and that for outside was simulated by using calculated seawater values at T-E1. The food chain transfer of <sup>137</sup>Cs to fish was simulated by 12 biological compartments with reported food web relation and food compositions (Tateda et al., 2013). The reconstructed levels in fish were verified with observed <sup>137</sup>Cs concentrations by TEPCO fish monitored concentrations. The radiocesium biokinetics in fish was evaluated, and determining factors were examined.

The simulated and observed <sup>137</sup>Cs concentrations in olive flounder is shown in Figure 1, because it was reported that the radiocesium level in olive flounder was well reconstructed in other coastal waters of the Fukushima accident affected area.

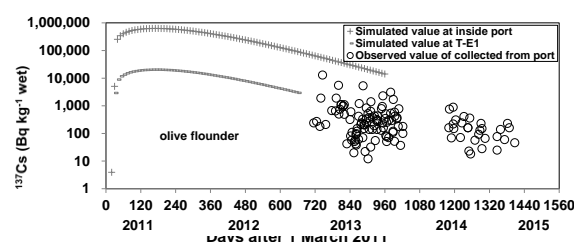


Figure 1. Simulated values and observed concentrations of <sup>137</sup>Cs in olive flounder inhabiting the vicinity of 1FNPP.

The observed <sup>137</sup>Cs concentrations in olive flounder collected from the port of 1FNPP were demonstrated as capped with the simulated values which used the observed <sup>137</sup>Cs concentrations of seawater in the port. The observed concentrations were similar to the simulated values for olive flounder using seawater concentration of T-E1. Thus large variation of <sup>137</sup>Cs concentrations in olive flounder collected from the port suggested that this species swam from the outside where it exposed to lower concentration of radiocesium of e.g. T-E1. The simulated maximum concentration of <sup>137</sup>Cs was theoretically derived as 620 kBq kg-wet<sup>-1</sup> at August 2011. However the actual level was presumed as lower than the model calculated attainable value, probably being 2 kBq kg-wet<sup>-1</sup> which was derived e.g. at T-E1. The ecological half-life ( $T_{eco1/2}$ ) of radiocesium for olive flounder was 290 d by using concentrations in collected fish at the port. This result was longer than the calculated  $T_{eco1/2}$  (170 d) for this species inhabiting other coastal waters of the Pacific Ocean along the eastern Japan. The result indicated that the radiocesium depuration in olive flounder inhabiting the vicinity of 1FNPP was probably delayed by food chain transfer.

## Acknowledgments

This work was supported by the Central Research Institute of the Electric Power Industry.

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# Development of $^{236}\text{U}$ -AMS in MALT

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Keywords: AMS,  $^{236}\text{U}$ , Tracer, Fukushima Daiichi nuclear disaster.

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$^{236}\text{U}$  is long lived isotope ( $T_{1/2} = 2.342 \times 10^7$  y) produced by thermal neutron capture of  $^{235}\text{U}$  in nature. However its natural abundance is so low that the natural isotopic ratio  $^{236}\text{U}/^{238}\text{U}$  is estimated to be the order of  $10^{-12}$ . Meanwhile  $^{236}\text{U}$  is produced much more artificially, by the nuclear bomb tests and in the nuclear reactor. Such anthropogenic  $^{236}\text{U}$  is transported by the global fallout and released by the spent nuclear fuel reprocessing plants into the environment. Consequently the  $^{236}\text{U}/^{238}\text{U}$  ratio has been increasing rapidly to the order of  $10^{-9}$ .

Recently,  $^{236}\text{U}$  has been recognized as a tracer in geochemical field. From the study of  $^{236}\text{U}$  depth profile in the Japan Sea,  $^{236}\text{U}$  seems to behave as a conservative nuclide in seawater and has potential advantages over other traces of oceanic circulation (Sakaguchi 2012).

For the detection of  $^{236}\text{U}$ , conventional mass spectrometry (inductively coupled plasma mass spectrometry: ICP-MS, thermal ionization mass spectrometry: TIMS) suffers from strong isobaric interferences of  $^{235}\text{UH}^+$ , which make the detection limits of the  $^{236}\text{U}/^{238}\text{U}$  to be  $10^{-7} - 10^{-10}$ . On the other hand, Accelerator Mass Spectrometry (AMS) enable to measure  $^{236}\text{U}/^{238}\text{U}$  of as low as  $10^{-13}$  (Steier 2010).

Aiming at the background level of  $10^{-13}$ , development of  $^{236}\text{U}$ -AMS system had started at MALT (Micro Analysis Laboratory, Tandem accelerator).  $^{236}\text{U}$  is extracted as  $^{236}\text{UO}^+$  from the ion source and exchanged to  $^{236}\text{U}^{5+}$  at the terminal of the accelerator. The magnetic rigidity at the injection magnet is  $5.07 [\text{MeV}^{1/2}\text{amu}^{1/2}]$ . The terminal voltage is 2.4 MV and the magnetic rigidity at the analyzing magnet is  $16.4 [\text{MeV}^{1/2}\text{amu}^{1/2}]$  (Figure 1.).

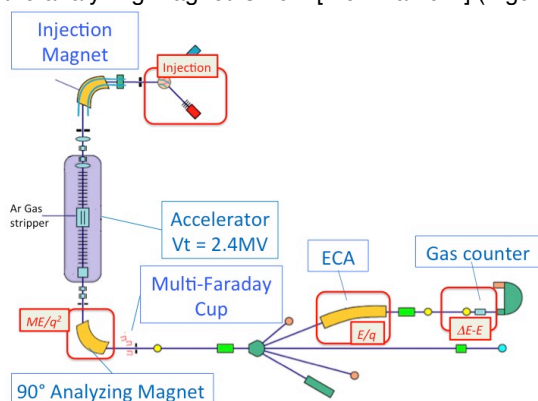


Figure 1. Schematic diagram of MALT beam courses.

The interference of  $^{235}\text{U}^{5+}$  coming from  $^{235}\text{U}^{16}\text{OH}^+$  was completely suppressed by inserting slits after the analyzing magnet and the ECA (Electrostatic Cylindrical Analyzer) (Figure 2.).

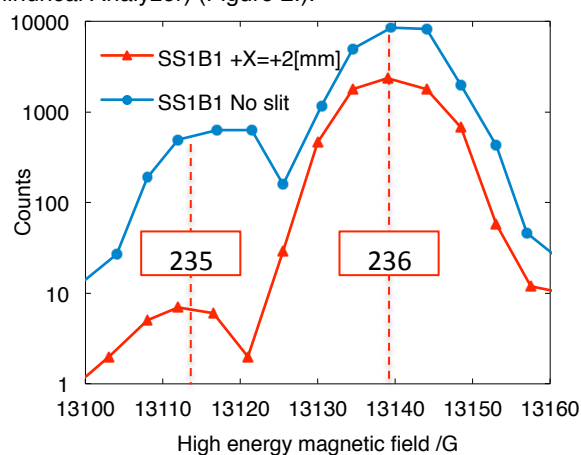


Figure 2. Position of slit after ECA and mass spectrum.

However another interfering component from the high rigidity side was found and determined the background corresponding to  $^{236}\text{U}/^{238}\text{U}$  of  $2 \times 10^{-9}$ . We conjecture that this interfering component is  $^{238}\text{U}$  which undergoes delayed charge exchange ( $^{238}\text{U}^{4+} \rightarrow ^{238}\text{U}^{5+}$ ) with residual gas during the second acceleration after the terminal resulting to have smaller energy. According to this hypothesis, the interfering process will be examined by both experimental and numerical method.

## Acknowledgement

The authors thank Prof. Peter Steier of VERA for providing standard samples.

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# Radiocesium distribution and fluxes in the typical *Cryptomeria japonica* forest at the late stage after the accident at Fukushima Dai-ichi Nuclear Power Plant

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Keywords: radiocesium, Fukushima, forest, radionuclide fluxes.

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As a result of the accident at the Fukushima Dai-ichi NPP, the large areas were contaminated with radiocesium, including 42,800 ha of forests with the level of contamination with <sup>134,137</sup>Cs above 1 MBq m<sup>-2</sup>, or about 66% of the total heavily contaminated area (Hashimoto *et al*, 2012). In differ to agricultural areas, where the large-scale decontamination has been successfully carried out, applicability of such a measure to the forest ecosystems is discussible, and Fukushima forests will likely remain contaminated for a long period.

Numerous studies revealed and quantified the key processes determining radiocesium redistribution (mainly its interception by the trees' crowns followed with the gradual removal to the soil profile with precipitations and litterfall) in Fukushima forests at the early stage after the accident (Kato *et al*, 2012; Kato and Onda, 2014; Teramage *et al*, 2014; Loffredo *et al*, 2014). However, at the later stage radiocesium redistribution in the forest ecosystems will be formed by the fluxes of its root uptake, return and internal translocation. Objectives of our study are characterization of the radiocesium distribution at the beginning of the late stage and parameterization of the above-mentioned fluxes for prognosis of the long-term behaviour of radiocesium in Fukushima forests.

For these purposes, in the spring of 2014 IER started the research project in the typical forest ecosystem (dominant tree species: *Cryptomeria japonica*, approx. 40-yr old artificial plantation, 335 trees ha<sup>-1</sup>) located in Yamakiya district (Kawamata town, Fukushima prefecture). Territory contamination density with <sup>137</sup>Cs in November of 2014 was 7.85·10<sup>5</sup> Bq m<sup>-2</sup>.

The experimental site was equipped for monitoring of the radiocesium biogenic fluxes (sampling of throughfall, stemflow and litterfall) and for assessment of the geochemical radiocesium migration flux in the root-inhabited soil layer based on recording of the moisture regime in the unsaturated zone. In the end of 2014, the sampling campaign was carried out for specification of the radiocesium distributions in the compartments of the aboveground biomass and in the soil profile and for characterization of the soil properties (bulk density, porosity, permeability etc).

Obtained results confirm that the stage of removal of the initially intercepted activity from the trees' crowns is almost over: in the end of 2014 only about 6% of the total radiocesium activity in ecosystem was found in the aboveground biomass, while litter and soil contained respectively 20% and 74%. Annual fluxes of radiocesium return to the soil surface with litterfall and throughfall were estimated as approx. 1% of the total

activity in the ecosystem each, while contribution of stemflow was two orders of magnitude lower. The litterfall flux decreased with time; however, comparable large fraction of radiocesium in the compartment of old leaves still may represent the residues of the initial deposition to foliage (Figure 1).

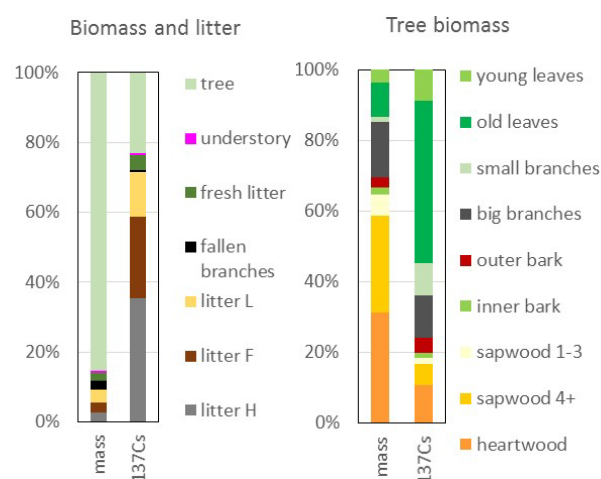


Figure 1. Distribution of <sup>137</sup>Cs in the biomass and litter compartments.

Our further studies in 2015 and 2016 will enable the accurate estimate of the root uptake and geochemical migration fluxes; the preliminary estimates of the fluxes are n% and n·0.1%.

## Acknowledgments

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# Radiocesium solid-liquid distribution, wash-off and migration within contaminated catchments after the accident at Fukushima Daiichi Nuclear Power Plant

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**Keywords:** Radiocesium; Fukushima; river basin; wash-off; migration

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A major earthquake and devastating tsunami on 11 March 2011 caused a nuclear accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), which resulted in contamination of wide range of terrestrial and freshwater environments by <sup>134</sup>Cs and <sup>137</sup>Cs. Contaminated territory of Fukushima Prefecture is characterized by differentiated hydrographic network, which includes the largest river of the area – Abukumagawa. All rivers finally enter to the Pacific Ocean. Catchments of the rivers contaminated because of FDNPP accident became a long-term source of secondary contamination of water bodies by surface runoff and radiocesium flux to the Ocean.

The purpose of this work is to assess Fukushima origin radiocesium mobility in the “soil-water” system based on the results of its monitoring in the rivers on the contaminated areas and using results of field experiments and observations.

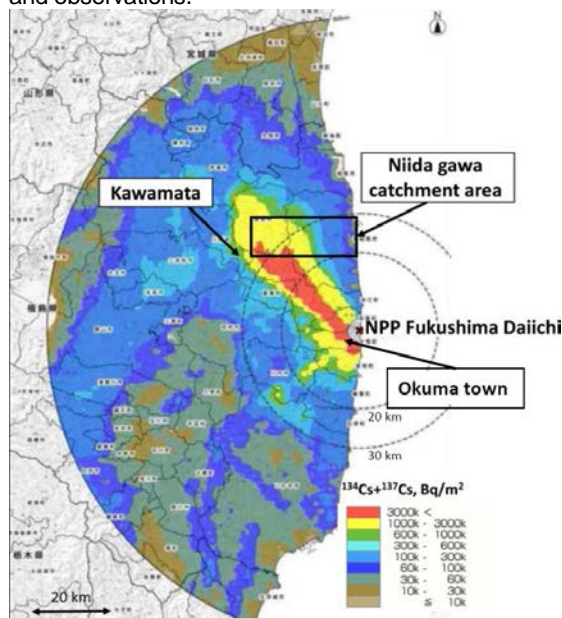


Fig. Map of the areas under study with radiocesium (<sup>134</sup>Cs + <sup>137</sup>Cs) deposition map according to the fifth airborne monitoring survey (MEXT, 2012) on the date of 28 June 2012.

Field observations were carried out in close-in area of the FDNPP – Okuma town (irrigation ponds and soils on their catchments), Niidagawa River basin and forest site in Yamakia (Kawamata town).

The effective dispersion coefficients in the Fukushima soils were found to be in range 2-10 cm<sup>2</sup>/year. Relatively high rates of radiocesium migration in Fukushima soils may be associated with high annual precipitation and high biological activity. In forest soils, the radiocesium dispersion is faster as compared to grassland soils more likely because of soil density difference. The study and analysis of the vertical distribution of the Fukushima origin radiocesium in the Niida river floodplain soils has made it possible to identify the rates of contaminated sediment accumulation on the floodplain. The average annual accumulation rate for sediments is quite non-uniform at different sections of the Niida river floodplain and varies from 0.3 to 3.3 cm/year depending on river bottom morphology, which is attributed to both the natural causes and human-induced impacts.

Taking into account the sediments accumulation leading to an increase in the radiocesium inventory in alluvial soils is key for predicting redistribution of radioactive contamination after the FDNPP accident on the river catchments, as well as for decision making on contaminated territories remediation and clean-up. Clean-up of alluvial soils does not seem to be worthwhile because of the following accumulation of contaminated sediments originating from more contaminated areas, including the exclusion zone.

$K_d$  values in the surface waters in the Fukushima area is high ( $10^5$ - $10^6$  L/kg), both for the river water and the surface runoff. This points to a high radiocesium binding capacity of sediments and soils in the Fukushima area and, as a result, a significant part of radiocesium on the area contaminated after the Fukushima accident are transported on suspended material with surface runoff and river flow.

Inversely, normalized dissolved wash-off coefficients  $N_i$  in the FDNPP area is low ( $\sim 10^{-4}$  m<sup>-1</sup>). Normalized particulate wash-off coefficients  $N_s$  received for the runoff plots and rivers of the Fukushima area are comparable. This suggest that the normalized radiocesium wash-off coefficients estimated on the runoff plots can be used for modeling and prediction of radiocesium transport to the Fukushima area rivers and beyond.

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# Assessment of residual doses to the population after the decontamination works in Fukushima prefecture

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Keywords: decontamination, residual dose, probabilistic approach, spatial and interpopulational variability, Fukushima Daiichi Nuclear Power Station accident.

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Large amount of radioactive materials were released to the environment by the Fukushima Daiichi Nuclear Power Station (1F) accident. Many residents in contaminated areas are exposed to radiation through their daily lives. Decontamination of buildings and living areas is one of the countermeasures for reducing the doses to the public in middle and long term situation. After the 1F accident, the decontamination criterion is fixed at 0.23  $\mu\text{Sv/h}$  in measured value of survey meters (Ministry of the Environment, 2011). This criterion was adopted to achieve the dosimetric goal of the annual effective dose of 1 mSv added by the radiation sources originating from the 1F accident (Nuclear Emergency Response Headquarters, 2011). The calculation of this criterion was made on the assumption that the people spend 8 hours outdoors and 16 hours indoors (sheltering factor: 0.4) in a day, and the contribution of natural radiation is 0.04  $\mu\text{Sv/h}$ . The objective of this study is to find out lessons learned concerning the decontamination criterion and its effects for a future planning of decontamination strategy. To achieve this, the following works were performed: (i) Reviews of the procedure to calculate the decontamination criterion after the 1F accident and to reveal insufficiency in it, (ii) Assessment of the extent of residual doses to the population living in Fukushima prefecture to analyze the effects of decontamination works that are performed in accordance with the dose criterion of 0.23  $\mu\text{Sv/h}$ .

As a result of the review of the procedure, we found out some issues that should be discussed at least. Firstly, a dose conversion is necessary to compare the readings measured by survey meters with the dosimetric goal expressed in the effective dose. Secondly, the contribution of natural radiation was determined from a national average. The calculation of criterion should be made using the values measured in Fukushima prefecture with consideration about a dose conversion. Thirdly, the same behavioural pattern was supposed to all populations without considering differences between occupations.

The residual doses were assessed using a probabilistic approach taking into account the issues mentioned above. The ambient dose equivalent, which means readings of survey meters, and the effective doses were calculated from Cs-137 deposited concentrations onto the ground with the dose conversions. Figure 1 shows the Cs-137 deposited concentrations for each municipality in Fukushima prefecture. These data were obtained from aerial radiation monitoring by Japan Atomic Energy Agency, which are decay-corrected to June 28, 2012. Geometric means and geometric standard deviations for the municipalities range from  $7.4 \times 10^{-1}$  to  $2.0 \times 10^3$  ( $\text{kBq/m}^2$ ), and from 1.0 to 3.9, respectively. The contribution of natural radiation sources was calculated based on the results of measurements in Fukushima prefecture just before the

accident with a conversion to the effective doses based on an empirical model reported by Moriuchi et al. (1989).

In our probabilistic assessment, the spatial variabilities of the Cs-137 deposited concentrations in Fukushima prefecture and the interpopulational variabilities of behavioural patterns for each occupation were considered. The data on behavioural patterns were obtained from our surveys actually performed in Fukushima prefecture. The three population groups of indoor workers, outdoor workers and pensioners were surveyed for the time to spend inside and outside houses, their work places and other activity places. From the results of a preliminary assessment for Minamisoma City, we found that the annual effective doses to the most of indoor workers and pensioners were below the dosimetric goal (1 mSv/y). In our presentation, we will report the details of our assessments of residual doses to each population group for other municipalities in Fukushima prefecture.

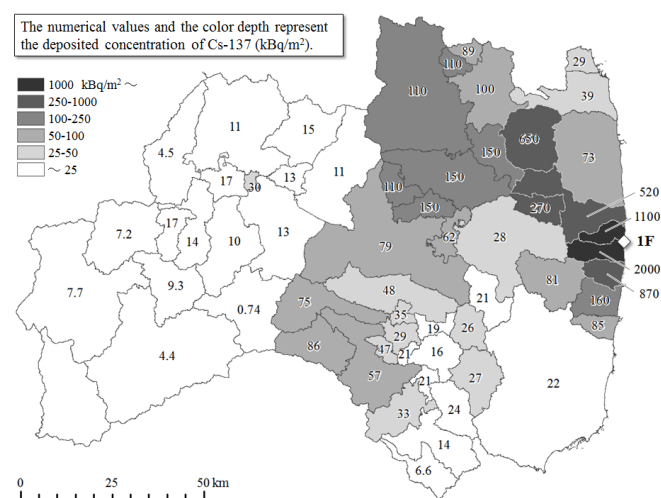


Figure 1. The geometric means of Cs-137 deposited concentration for each municipality in Fukushima prefecture.

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# Radioactive characterization of residues from an Oil industry in Ghana

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Keywords: scales, NORM industry, uranium, thorium

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During the last decade some oil explorations have started at different sites along the west coast of Ghana. Consequently, environmental concern has appeared in association with possible chemical pollution but, in addition, radiometrical characterization becomes relevant as far as oil extraction is concerned, is a well-known NORM (Naturally Occurring Radioactive Material) activity.

Residues in the oil industry could accumulate natural radionuclides and hence, contribute to internal/external doses in humans, particularly during production, maintenance, the transport of waste and also during the decontamination works. Furthermore, environmental concern appears regarding places where processing and disposal of waste take place. Exposures of a similar nature may also arise during the decommissioning of oil production facilities.

To evaluate the levels (activity concentration) of natural radionuclides in residues from an oil industry in Ghana, several samples (scales and sludges) were sampled in December 2014 from the Jubilee oilfield at the West Coast. Direct external gamma dose rate measurements were performed, dose rates found were in the range of up to a few  $\mu\text{Sv/h}$ . In exceptional cases, dose rates measured directly on the outside surfaces of production equipment have reached several hundred  $\mu\text{Sv/h}$ , which is about 1000 times greater than normal background values.

With basis in these previous results, radiometrical measurements were performed via alpha spectrometry with PIPS detectors ( $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  isotopes) and gamma spectrometry with a high resolution HpGe detector ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{224}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{234}\text{Th}$  and  $^{40}\text{K}$ ) in sludge and scale samples. Also elementary characterization via ICP-MS and SEM (Scanning Electron Microscopy) were applied within this set of samples.

Results in sludges via alpha spectrometry show a negligible contribution both in U and Th isotopes with activity concentrations around or below 0.01 Bq/g. Regarding gamma measurements (Table 1) higher activities concentration were obtained (mainly associated to Ra isotopes) but still below the *Exemption levels* (EL) according to recommendations in the IAEA BSS 1996. The elementary composition in these sludges is mostly Fe and Cu but also Ba.

Table 1. Activity concentration via gamma in sludges. Between brackets are expressed the uncertainties in the determinations. EL\* *Exemption levels*

Sample	$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{228}\text{Th}$	$^{40}\text{K}$
Mean	0.73 (0.24)	0.46 (0.23)	0.17 (0.05)	0.05 (0.01)
Range	0.56- 0-90	0.30- 0.82	0.13- 0.20	0.04- 0.06
EL*	10	10	1	100

Regarding scale samples,  $^{238}\text{U}$  activity concentration ranges from 0.22-0.25 Bq/g and  $^{232}\text{Th}$  0.04-0.07 Bq/g, also below EL. However (Table 2)  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and their respective progenies, have much higher activity concentrations exceeding the exemption level.

Table 1. Activity concentration via gamma in scales. Between brackets are expressed the uncertainties in the determinations. EL\* *Exemption levels*

Sample	$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{228}\text{Th}$	$^{40}\text{K}$
Mean	37.1 (1.3)	29.8 (0.8)	11.0 (0.8)	2.0 (0.01)
Range	36.2- 38.0	29.4- 30.2	10.4- 11.8	1.9- 2.1
EL*	10	10	1	100

Then, one important conclusion in this work is that scales management requires supervision by workers with radiation safety training.

## Acknowledgments

The authors would like to thank the IAEA for the financial support and also to Tullow Ghana Staff for their cooperation and assistance.

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# Mobility of radium in phosphogypsum waste generated by the wet sulphuric pathway

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Keywords: phosphogypsum, radium, mobility, BCR sequential extraction.

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## Introduction

Phosphogypsum (PG) is a waste generated in the production of phosphoric acid by the sulphuric acid route, which phosphate rock ( $\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$ ) is subjected to a chemical treatment with sulphuric acid ( $\text{H}_2\text{SO}_4$ ). The sedimentary phosphate ore usually contains high concentrations of natural radionuclides, and finally a significant fraction of them remain in the PG. In the southwest of Spain, PG has been stored in piles on wetlands of Huelva estuary for 45 years. Currently, those piles reach 5 m of height, covering an area of about 1000 ha. It is estimated that there are over  $10^8$  t of PG are stored in these stacks, which involve potential radiological and environmental risks. (Bolívar et al., 2009; Pérez-López et al., 2010).

The main purpose of this paper is to apply the optimized sequential extraction procedure BCR (Community Bureau of Reference) (Rauret et al., 1999), to evaluate the mobility of the radium in PG.

## Methodology

In this study, a PG core up to reach the base of wetland was taken. The BCR method is based on three-step extraction, which uses different extractants with different chemical aggressiveness (Table 1). Each fraction was subjected to a sequential radiochemical procedure to isolate the Ra, and then electrodeposited onto stainless steel discs. Subsequently and finally is counted by alpha-particle spectrometry with PIPS detectors.

Table 1. Sequential extraction procedure BCR.

Fraction	Extractant	Operation Conditions
F1	0.11M $\text{CH}_3\text{COOH}$	Stirring for 16 h
F2	0.5M $\text{NH}_2\text{OH} \cdot \text{HCl}$	Stirring for 16 h
F3	8.8 M $\text{H}_2\text{O}_2$ +1M $\text{NH}_4\text{OAc}$	Digestion for 1 h at AT; digestion for 1 h at 85 °C; and stirred for 16 h
RF	Aqua regia	Digestion for 16 h at AT; Digestion at ET for 2 h.

The sum of the three-step sequential extraction (MF = F1+F2+F3) will correspond to the total content of mobile radioelement, and therefore this MF fraction could be released into the environment. The residual fraction (RF) contains elements associated strongly to crystal structures of minerals, and this RF is unlikely to be released.

## Results

The PG contains from 400 to 900  $\text{Bq kg}^{-1}$  of  $^{226}\text{Ra}$  and does not present any trend in depth. At the same way, the radium contents in each fraction present a high dispersion. This fact is probably related to the origin of used ore to produce phosphoric acid for the 45 years of active industrial production

By considering the conditions at the PG is exposed (rainy winters and hot and dry summers), the most polluting fractions to the environment are interchangeable (F1) and oxidizable (F3).

This study concludes that the mobile fraction of radium could represent up to 70% of total content  $^{226}\text{Ra}$  in the PG. This fact implies in future the possibility of big Ra releases into the aquatic environment depending of the environmental conditions.

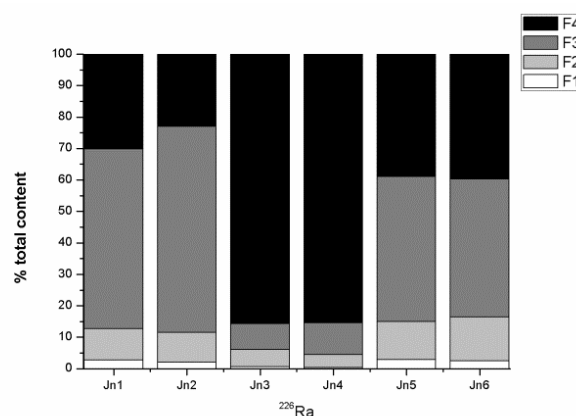


Figure 1. Percentages of  $^{226}\text{Ra}$  extracted from each fraction in the phosphogypsum samples.

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# Occupational exposures in two industrial plants devoted to the production of ammonium phosphate fertilizers

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Keywords: occupational exposures, phosphate industry, external radiation, aerosol filters, radon

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## Introduction

The largest industrial chemical complex in Spain is located in the vicinity of the city of Huelva, in the estuary formed by the confluence of the Odiel and Tinto river mouths. As a part of this complex, there is a NORM industry devoted to phosphoric acid production, and as consequence to the production of ammonium phosphate fertilizers from the phosphoric acid generated.

When these studies were performed, four plants devoted to the production of phosphoric acid (with a nominal production for each plant of  $2.0 \times 10^5$  t of  $P_2O_5$  per year), two plants devoted to the production of mono-ammonium phosphate (MAP, nominal production of  $2.7 \times 10^5$  t of MAP per year) and one plant devoted to the production of di-ammonium phosphate fertilisers (DAP, nominal production of  $2.7 \times 10^5$  t of DAP per year) were in operation, thereby forming the largest industry in southern Europe devoted to the production of phosphoric acid and ammonium phosphate fertilizers.

## Aim and Methodology

According to the international regulation affecting NORM industries, assessments of occupational exposure should be performed in all the plants devoted to the production of phosphoric acid and derived phosphate fertilisers in order to ascertain whether any actions or radiological protection measures should be taken for the mitigation of the effective doses to which the workers are susceptible. In this sense, we have evaluated the occupational exposures in one of the twin plants devoted to the production of MAP fertilisers and in the plant devoted to the production of DAP fertilisers. In both places, the determination of external gamma dose rates was performed using a portable dosimeter at several locations inside the plants, aerosol filters were collected for evaluation of the exposures due to inhalation of particulate matter and  $^{222}\text{Rn}$  concentrations were determined by using passive detectors.

## Results

### External gamma exposures

A detailed map of these gamma dose rate increments over background (which is  $0.09 \mu\text{Sv h}^{-1}$ ) was created for each of the two analysed plants and it was observed that the values were low and always in the range  $0\text{--}0.08 \mu\text{Sv h}^{-1}$ .

### Committed effective doses due to inhalation

The radiometric determinations performed in the aerosol filters collected in the plants show enhanced (and variable in time) mass activity concentrations of the radionuclides belonging to the uranium series in relation to the concentrations determined in the background area. However, no enhancements were observed for the radionuclides from the thorium series or for  $^{40}\text{K}$ .

The committed effective doses calculated by a conservative way with the activity concentration values obtained for the different worker categories show that in all cases remain below the value of  $0.12 \text{ mSv}$ .

### $^{222}\text{Rn}$ in the plants

On the other hand, the  $^{222}\text{Rn}$  determinations performed in the MAP and DAP facilities by placing a couple of passive detectors (CR-39 type) in each plant have yielded the following values:  $18 \pm 3$  and  $16 \pm 3$  for MAP and  $15 \pm 3$  and  $18 \pm 3 \text{ Bq m}^{-3}$ , for DAP, where the uncertainties are expressed as the standard deviation of the mean.

## Conclusions

A detailed radiological study, performed in a couple of NORM plants devoted to the production of ammonium phosphate fertilisers (MAP and DAP), located in Huelva (south-west Spain), has allowed us to conclude that the occupational doses received by the different worker categories do not reach the reference value of  $0.3 \text{ mSv yr}^{-1}$ , considering the worst scenarios and all the possible pathways of exposure. The dose increments due to the external radiation and inhalation of particulate matter are the main routes of exposure. No single action or adoption of any radiation protection measure needs to be taken in order to protect the health of the workers. Furthermore, the  $^{222}\text{Rn}$  concentrations inside the two analyzed plants are also of no concern since they are around  $20 \text{ Bq m}^{-3}$ , which is one order of magnitude lower than the established limit that would trigger the adoption of remediation actions.

## Acknowledgments

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# NORM measurements at the beach sand and coastal sediments near a mining area, Ierissos Gulf, Greece

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Keywords: Stratonis, NORM, mine industry, coastal environment.

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From the anthropogenic activities special care must be given to mining, as may not only affect the soil locally but also have a wider impact to the aquatic ecosystems due to mining-milling operations, the acid mine drainage, the erosion of waste dumps and the disposal of tailings (Razo et al., 2004). The types of radioactive materials generated from the mining of the ores include  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and their progenies (NORM: Naturally Occurring Radioactive Materials). One of the active mining activities of Greece is located in Stratonis (Ierissos Gulf, North Greece) and the mining activity has a long history, with mining and metallurgical peak in 1970's. The monitoring of the region was held primarily from the exploration company (Kelepertzis et al. 2012), emphasizing mainly to the terrestrial area and to heavy metal environmental status.

In this work the major motivation was to provide a baseline information of the studied coastal area concerning the radioactivity levels of the seabed. Additionally, to investigate possible correlation among mobility of radionuclides taking into account their accumulation in beach sands and coastal sediments. For this effort a line-mapping measurements along the coast were performed taking under consideration the distance from the load-out pier area and river estuaries. So samples of beach sand, coastal sediment and seabed sediment were collected for further analysis.

A total of 30 samples were collected along the coast of Ierissos Gulf in 6 different locations (Str 1, 2, 13, 14, 15, 16). For each location two samples (beach sand (Str#\_b) and coastal sediment (Str#\_a)) were collected. In addition, at the sampling points of the load out pier area and the reference point, sediment samples of the seabed were collected, with code names Str1 and Str16 respectively. All the samples were prepared at the Institute of Oceanography of the Hellenic Centre for Marine Research (HCMR). The samples measured via gamma-ray spectroscopy were processed by a standard procedure including drainage, sieving and pulverization and were tightly closed to their final boxes for at least 21 days in order to achieve secular equilibrium between Radon daughters ( $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ) and  $^{226}\text{Ra}$ .

The natural radionuclides in marine sediments were analysed using a 100% (nominal relative efficiency) High Purity Ge detector (HPGe). The energy and efficiency calibration were performed using  $^{152}\text{Eu}$ ,  $^{40}\text{K}$  and  $^{238,235}\text{U}$  sources as is described in Ref. (Eleftheriou, 2014).

The activity concentrations of  $^{238}\text{U}$  series are depicted for all stations in Fig.1. More specifically, the

activity concentrations of  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  were found to range between  $(8 \pm 1)$  Bq/kg and  $(100 \pm 7)$  Bq/kg. Furthermore, the activity of  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$  and  $^{208}\text{Tl}$  varied from  $(11 \pm 1)$  Bq/kg to  $(180 \pm 7)$  Bq/kg. Concerning  $^{40}\text{K}$  and  $^{210}\text{Pb}$ , their activities varied from  $(320 \pm 33)$  Bq/kg to  $(1100 \pm 109)$  Bq/kg and from  $(23 \pm 7)$  Bq/kg to  $(150 \pm 15)$  Bq/kg, respectively. The highest concentrations were observed in the estuary of Kalatzi Lakos River (Str 15\_0a).

For each sampling location, the activity concentrations of beach sand samples and the coastal sediment ones were similar within uncertainties. Furthermore, activity concentrations of the sampling points near the load out pier area (Str 1, 2) are 3 times higher than the ones of the reference point (Str 16). Moreover, the activity concentrations of the area's river estuaries (Str 13, 14, 15) are in the same level as the ones of the reference point, with exception of the coastal sediment sample of Kalatzi Lakos River (Str 15\_0a).

Further investigation is being held in the seabed near Stratonis port (where the load out pier area is), as well as along the length of the two main rivers of Ierissos Gulf (Kokkinolakos River and Kalatzi Lakos River).

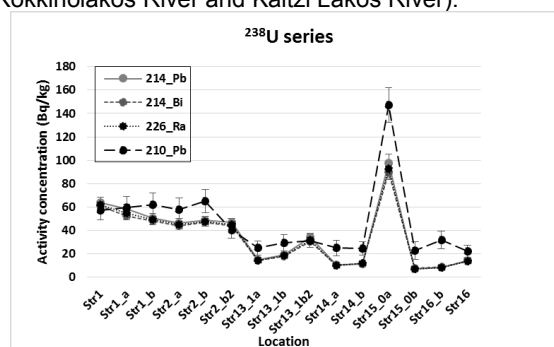


Figure 1.  $^{238}\text{U}$  series activity concentrations.

## Acknowledgments

Ms F.K. Pappa would like to thank A.G. Leventis Foundation for supporting this research project.

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# Transfer of Ra-266 from soil contaminated with U-mill tailing to radish (*Raphanussativus L.*) and savoy (*Brassica olerancea L. var. sabauda*)

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Keywords: Ra-266, transfer, plants

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During the operation of the former uranium mine Žirovskivrh, Slovenia in 80s, mining and milling wastes were deposited on nearby waste piles. The wastes contain elevated levels of natural radionuclides from the uranium decay chain which can be with different migration processes (erosion, aerial deposition, groundwater) discharged into the environment and taken up by plants. Accumulation of radionuclides by plants growing on soil contaminated with uranium mill waste was already reported by several authors (Soudek et al., 2010, Petrova, 2006, Štok et al., 2011). Members of Brassicaceae family are known as plants with higher ability to accumulate heavy metals as well as radionuclides (Vera Tomé et al., 2009). Therefore Brassica crops are promising candidates for the phytoremediation but on the other hand, caution is needed when they grow on contaminated site to prevent transfer of radionuclides to food chain.

The aim of our study was to investigate accumulation of Ra-266 in radish (*Raphanussativus L.*) and savoy (*Brassica olerancea L. var. sabauda*). Various contamination levels were used in a pot experiment (Fig. 1) to evaluate transfer of Ra-266 from the soil contaminated with uranium mill to the radish and savoy. After growing season, Ra-266 in contaminated soil and above ground parts was analysed. Measurements were performed by gamma-ray and alpha-particle spectrometry. Finally, the transfer factors were calculated as ratio of average activity concentration in plant samples and average activity concentration in soil for different contamination levels.

The obtained activity concentrations of Ra-226 in radish were  $9.36 \pm 7.04$ ,  $68.1 \pm 6.4$ ,  $57.8 \pm 13.6$ ,  $55.1 \pm 17.4$  and  $115 \pm 9$  Bq kg<sup>-1</sup> dry mass for control, 20, 40, 60 and 80 % of uranium mill tailing content, respectively. On the other hand the activity concentrations in savoy were  $21.0 \pm 7.4$ ,  $31.7 \pm 9.4$ ,  $35.4 \pm 14.6$ ,  $41.2 \pm 10.7$  and  $61.9 \pm 9.0$  Bq kg<sup>-1</sup> dry mass for control, 20, 40, 60 and 80 % of uranium mill tailing content, respectively.

The obtained results show the highest Ra-266 activity concentration for plants growing in

substrate with the highest uranium mill content. The lowest transfer factors were for the highest uranium mill content due to the significantly higher activity concentration. Radish could be potentially used for phytoremediation due to higher uptake of Ra-226 compared to savoy.

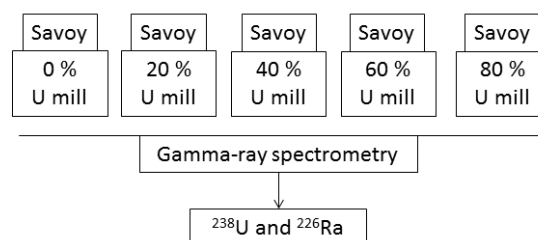


Figure 1. Scheme of pot experiment.

## Acknowledgments

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# Natural Radioactivity of Building Materials in Greece

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**Keywords:** gamma spectroscopy, building materials, index factor.  
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Most individuals spend 80% of their time indoors and natural radioactivity in building materials is a source of indoor radiation exposure. Indoors-elevated dose rates may arise from high concentrations of radionuclides in building materials, from both internal and external exposure. The external exposure is caused by direct gamma radiation and in dose calculations, the main radionuclides of interest are Ra-226, Th-232 and K-40.

The guidelines of the European Commission Radiation Protection 112 are the first comprehensive document issued by the EC, which sets the principles of radiological protection concerning natural radioactivity (both external and internal) of building materials. The new European Basic Safety Standards Directive (BSS) sets down a framework for controlling exposures to natural radiation sources arising from work activities. The purpose of setting controls on the radioactivity of building materials is to limit the radiation exposure due to materials with enhanced or elevated levels of natural radionuclides. Since 2006 the Greek Atomic Energy Commission has issued a Decision, adopting RP-112, to regulate the re-use of NORM by-products in construction of buildings and roads and the import and use of building materials that may contain elevated NORM concentrations.

Typical by-products used in the building material industry are: fly ash and bottom ash, alum shale, and phosphogypsum. Fly-ash and bottom-ash (slag) is reused from coal-fired power plants. Fly-ash is mainly used in the production of cement and sometimes in the production of other building materials such as bricks. In Greece, according to the Concrete Technology Regulation, fly ash is used as an additive to cement production up to a maximum percentage of 50%, and

cement, in turn, is used in concrete up to a maximum percentage of 13%.

The Department of Environmental Radioactivity Monitoring uses gamma-spectroscopic analysis techniques for the determination of Ra-226, Th-232 and K-40 in building materials. The concentration of Ra-226 is calculated based on the direct determination of Ra-226 and U-235 from the analysis of the multiplet photopeak at 186 keV. The index *I* is used for screening building materials of radiological concern.

$$I = \frac{Ra226}{200} + \frac{Th232}{300} + \frac{K40}{3000}$$

The scope of this work is to present the results of the measurements of building materials conducted in the laboratory of our Department in the time period 2010-2014, determine the average radioactivity content, and perform *I*-index calculations. More specifically, materials include fly ash samples used in the production of cement in Greece, wood, tiles and granites. The results of this work are compared to the findings of other studies regarding activity concentrations of building materials in the EU (Trevisi et al., 2012).

## Acknowledgments

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# Characterization of scales from a NORM (TiO<sub>2</sub>) Industry

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**Keywords:** Scales, NORM industry, Uranium, Thorium  
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The TiO<sub>2</sub> industrial process uses the mineral ilmenite as raw material containing natural radionuclides from <sup>232</sup>Th series (0.04-2 Bq/g) and <sup>238</sup>U ones (0.03-0.4 Bq/g). Previously [Cooper et al, 1981] found in one Australian TiO<sub>2</sub> facility values of activity concentrations in the range of 1644 Bq/g in <sup>228</sup>Ra or 415 Bq/g in <sup>226</sup>Ra in some samples from the hydrolysis area. For that reason, this industrial activity is in the IAEA list of typical NORM industries.

During the radiometrical study carried out in a TiO<sub>2</sub> pigment factory placed in the south west of Spain, external gamma measurements were performed along all the facility. A background value of 0.08-0.09  $\mu$ S/h [Bolivar et al. 2011] was measured outside the facility, but several values at the hydrolysis area (up to 1.10  $\mu$ S/h) and some pipes at the crystallization site (up to 12.60  $\mu$ S/h) pointed out “hot spots” at that facility. In the hydrolysis area, the high values observed were related with the concentration of radionuclides in some filters known as “Moore filters” while the high values in some pipes was due to the presence of scales. A scale is a coating or incrustation layer formed by mineral precipitation (initially in solution form) that occurs at the inner side of tubes or pipes in some chemical industries.

In this work, several scales at different pipes and one Moore filter were sampled from this plant. The main aim was to perform the characterisation of these “hot spots” not only from a radiometric viewpoint but also through its elementary composition. For that purpose, the following techniques were applied: alpha spectrometry (U, Th isotopes), gamma spectrometry with HpGe detectors, X-Ray fluorescence (XRF), Scanning Electron Microscope (SEM) and X-Ray Diffraction (XRD).

Regarding the Moore filter (original size 1.60 x 2.14 m), a sample of 0.8 x 0.8 m was measured via gamma founding 20 kBq/m<sup>2</sup> (<sup>228</sup>Th and <sup>228</sup>Ra), 8-9 kBq/m<sup>2</sup> (<sup>226</sup>Ra) and 1 kBq/m<sup>2</sup> of <sup>40</sup>K (all these values with 10% total combined uncertainty at 1-sigma criteria). By SEM (see Figure 1) we have obtained the following elemental information: 87.4% in atomic weight of C and O but also 6.0 % Ti, 4.1% S, 1.98 %Ba, 0.4% Pb, 0.06% Ca, 0.05% Sr and 0.05% Fe. The trapped “hot particles” in the filter contains Ca, Sr and Ba and is expected to have also Ra (measured via gamma), but with really very low concentration (below parts per billion, ppb). A wider result with XRF

technique (with a 2 x 2 cm filter) confirms the same elements but with another proportions that the ones shown before.

In relation with the scale samples, after their radiometrical characterization, we can indicate that in all the cases were found activity concentrations much higher than 1Bq/g for all the radionuclides from the U and Th series. So, from a radiological point of view, some actions need to be implemented for the protection of workers at these sites (specially during cleaning/dismantling of these sites).

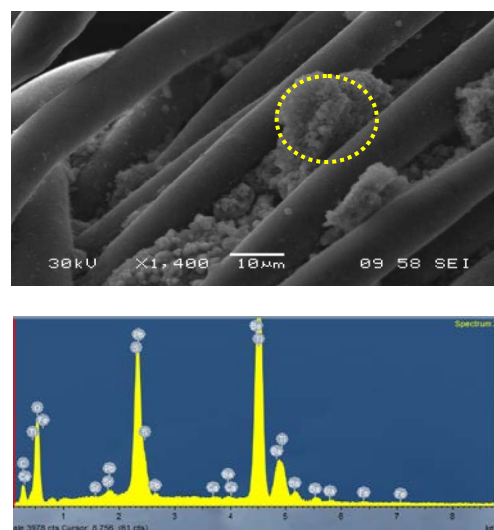


Figure 1 (Up). SEM picture (secondary electron image) from trapped particles in a Moore filter. (Down) X-ray spectrum belonging to a 20  $\mu$ m spot size over the area marked with a yellow circle in the picture.

## Acknowledgments

This work was financed by Spanish Nuclear Security Council (CSN) through the project “Radiological Impact associated with Non-Nuclear Industries from South of Spain”.

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## Simple analytical models of radionuclide transport in topsoil

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*Keywords: radionuclides, migration, soil profile, transport model.*

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A number of simple analytical models is available, which have been used to describe the vertical distribution of radionuclides originating from fallout, such as <sup>137</sup>Cs or <sup>90</sup>Sr and plutonium isotopes for anthropogenic, and <sup>210</sup>Pb or <sup>7</sup>Be for natural radionuclides, in the top layers of undisturbed soils. Fallout history can be of almost pulse type, such as for the cases of nuclear accidents, long lasting, as for global fallout, or continuous as for natural radionuclides.

Some of these models are merely empirical or descriptive, others based on physical and chemical laws which control transport and migration in porous media (such as soil), and interaction with soil matter by retention and sorption. The latter models are being developed not only, or not in the first place for describing a profile, but for predicting it in the future.

Among the descriptive models is the very popular simple exponential one and models from the family of Gaussian shaped functions. Possibly the most important physical model relies on the convection-diffusion

equation (CDE) (e.g. Bossew and Kirchner 2004). Others are based on probabilities of sorption, retention and remobilisation of tracers.

These analytical models which are solutions of transport equations include severe simplifications, such as assumption of temporal and spatial constancy of migration parameters, simplified sorption models etc. Nevertheless, they often perform surprisingly well.

We discuss several models, address their limitations and compare their performances. Empirical data of vertical profiles of radionuclide concentrations in soils contaminated by Chernobyl and Fukushima fallout are used to demonstrate the capability of such models.

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## Plutonium and uranium isotopes in Mongolia surface soils

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Radionuclides and their isotope ratios have been used as a tracer of environmental behaviors of elements (Hirose, 2011). Especially plutonium and fission products in aerosol and deposition samples before the FDNPP accident, whose major sources are dust blown by severe storms in the deserts and arid regions, are recognized as a proxy of environmental change due to human activities (Hirose et al., 2003). Plutonium has been released into the environment due to the atmospheric nuclear-weapons tests, burn-up of nuclear-battery satellite, nuclear reactor accidents, and others. Plutonium deposited on land surface, which is one of chemically reactive elements, is cycled in the globe according to natural processes. Effectively to use radionuclides as an environmental tracer, it is important to elucidate geographical distributions of radionuclides in continental land surface. In this paper, we describe the first sight of plutonium and uranium isotopes in surface soils of the southeast Mongolia, where corresponds to downwind regions of Lop nor nuclear test site and is a potential source region of continental dust affected in the northern hemisphere.

Undisturbed surface soil samples were collected by using a core sampler in southeastern Mongolia in fall 2007 (Igarashi et al., 2011). After sample preparation treatments, the fraction that passed through a 53- $\mu$ m mesh (nominal) was subjected to radioactivity analysis. 24 samples were used for plutonium analysis. The soil samples were decomposed and dissolved by strong acids. After the radiochemical separation and purification (Otsuji-Hatori et al., 1996), plutonium isotopes were measured by  $\alpha$ -spectrometry. Ten soil samples were subjected to the uranium isotope measurement. For the measurement, the samples were first subjected to the acid leaching treatments, and then the  $^{235}\text{U}/^{238}\text{U}$  ratios in the leachate (acid soluble part) and the residue (acid insoluble part) were measured by ICP-QMS separately. The obtained  $^{235}\text{U}/^{238}\text{U}$  ratios in the samples were corrected by that in seawater as a conventional standard, whose isotope ratio is regarded as the natural ratio.

The concentrations of  $^{239,240}\text{Pu}$  in Mongolian surface soils (24 samples) ranged from  $0.42\pm 0.03$  to

$3.53\pm 0.09$  mBq g<sup>-1</sup> with an average of 1.59 mBq g<sup>-1</sup>. The level of  $^{239,240}\text{Pu}$  in Mongolian surface soils was about one order of magnitude greater than that in Tsukuba, Japan, ranging from  $0.019\pm 0.005$  to  $0.41\pm 0.03$  mBq g<sup>-1</sup> as an average of 0.18 mBq g<sup>-1</sup>, although soil samples collected in Tsukuba contained ones in cultivated areas. The  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity ratio is a kind of fingerprint to identify sources of plutonium in the environmental samples. The  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity ratios in the Mongolian surface soil were in the range of 0.013 to 0.06 with an average of 0.029. The  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity ratios coincide with that in global fallout (0.03). These finding suggests that plutonium in the Mongolian surface soils is primarily derived from global fallout. However, it is further issue why the higher plutonium concentrations occurred in Mongolian surface soils, although lower precipitation amounts have been observed in Mongolia. The  $^{235}\text{U}/^{238}\text{U}$  ratios in both the acid soluble and acid insoluble parts of the measured soil samples were regarded as the natural ratio within analytical uncertainty, although the leaching proportion of uranium varied sample by sample widely. In addition, no dependence of the uranium isotope ratio on the particle size of soil was observed.

### Acknowledgments

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# Mass spectrometric measurement of Uranium isotope ratios in nuclear accident contaminated soil samples

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Keywords: Uranium, Fukushima, MC-ICPMS, Soil  
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Environmental contamination with alpha-emitting nuclides of uranium and transuranium elements are possible during accidents involving nuclear devices or nuclear power plants. Uranium comprises three natural isotopes viz.  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  with relative isotopic abundances of 0.0054%, 0.720% and 99.275%, respectively. Since soil samples contain high concentration of inherent natural uranium, it is more difficult to detect small quantities of U from man made nuclear sources. Therefore, accurate measurement of uranium isotopic composition in environmental samples is important to estimate the origin of nuclear materials and act as isotopic fingerprints.

For the measurement of isotopic fingerprints in the environmental sample, thermal ionization mass spectrometry (TIMS) is well established for the accurate and precise measurement of uranium ratios and is the reference method for the certification of many analytical standard materials. Since late 1990s, multi-collector inductively coupled plasma mass spectrometry has been introduced due to high ionization efficiencies and relatively short measurement time with great success competing with TIMS. A single focusing magnetic sector multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS) (IsoProbe; GV Instruments Ltd., Manchester, UK) has been used for the measurement of uranium isotopes (Watanabe and Nakai 2006).

An accurate and simple analytical technique for chemical purification of U in contaminated soil samples was developed with slight modification (Takamasa et al. 2013) used for sulfide samples. Since Japanese soil samples contain relatively large amounts of Fe, UTEVA resin was used twice. The total blank throughout the analytical procedure was ~ 9 pg for U.

In this study, we selected eight soil samples and two litter samples in close proximity to Fukushima daiichi nuclear power plant accident (FDNPP), a soil sample from Chernobyl nuclear power plant accident (CNPP) with high Cs-137 activity (Mishra et al. 2014) and one soil from Kosovo in depleted uranium (DU) conflict area. We also measured uranium isotopes from two Japanese soil samples before FDNPP accident as global fallout. Measurement of  $^{234}\text{U}/^{238}\text{U}$  and  $^{235}\text{U}/^{238}\text{U}$  in all samples were carried out, using a MC-ICPMS and plotted a graph (Fig. 1) between  $^{234}\text{U}/^{238}\text{U}$  and  $^{235}\text{U}/^{238}\text{U}$  in Fig. 1 to show the difference among DU, enrichment and natural uranium.

We will discuss in detail isotopic composition of uranium in soils contaminated by Chernobyl and Fukushima accident with fallout to demonstrate the viability of such method of analyses.

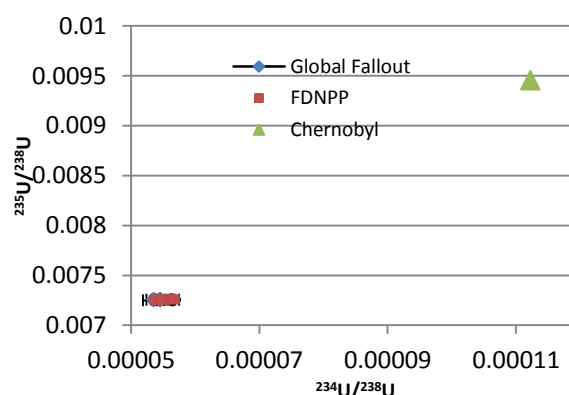


Figure 1.  $^{234}\text{U}/^{238}\text{U}$  vs.  $^{235}\text{U}/^{238}\text{U}$  to show differences between enriched U and global fall out

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## <sup>226</sup>Ra in Belgian soils: synthesis of student works at ISIB

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Keywords: natural radioactivity, radium 226, soil, radon  
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The experimental study of natural radioactivity is an important part of the study programme in physical and nuclear engineering at ISIB, Brussels. We report here on series of measurements of <sup>226</sup>Ra activity in soil. About 100 data were included in student theses and in the reports of the annual international intensive course XIMER. Due to the scarcity of other data for <sup>226</sup>Ra in soil, this dataset is an important contribution to the knowledge of natural radioactivity in Belgium.

<sup>226</sup>Ra was measured by high-resolution gamma spectrometry of soil samples generally taken at 1 m depth. A few samples were taken at a lower depth, from 0.5 m to 0.8 m, when the presence of a hard bedrock did not allow sampling at 1 m. The samples were dried, crushed, mixed with 10% charcoal and put in 250 ml bottles during one month for establishing the secular equilibrium between <sup>226</sup>Ra and <sup>222</sup>Rn progeny. The sample spectra were compared to a <sup>226</sup>Ra calibrated standard in the same geometry.

The sampling sites were georeferenced. They are distributed in Brussels and its surroundings, and in the Walloon region, discarding most of the Flemish region. The sampling is not statistically representative, because each small campaign was focused on a specific soil type or geological context.

The data are analysed with respect to the soil textural class and to the geological context. They are compared with another published dataset established in collaboration by two research institutions IHE and SCK-CEN (Deworm et al. 1988). These 35 data cover the whole Belgian territory with a low density, the samples being collected at 0.3 m depth.

Table 1 summarizes the results considering the geological context. Table 2 considers the soil texture. The textural classes adopted for the Belgian soil maps are:

U: heavy clay  
E: light clay  
A: loam  
L: sandy loam  
P: light sandy loam  
S: loamy sand  
Z: sand  
G: stony loam

Sampling sites with uncertain geological context or textural class were discarded.

The ISIB dataset also includes 28 measurements of soil permeability and <sup>222</sup>Rn in soil air at 1 m depth, in Quaternary loess and Tertiary sand.

Table 1. <sup>226</sup>Ra activity in soil (Bq/kg) according to the geological context. N=number of data.

Geology	ISIB mean (N)	IHE-SCK mean (N)	range
Quaternary loess	41 (35)	45 (6)	31-55
Tertiary sand	14 (11)	18 (10)	6-33
Tertiary clay	-	20 (3)	14-23
Alluvia	42 (5)	-	36-50
Mesozoic	-	26 (3)	21-29
Dinantian	37 (7)	-	28-47
Upper Devonian	29 (1)	43 (3)	28-56
Middle Devonian	-	35 (2)	31-38
Lower Devonian	35 (11)	31 (2)	22-48
Ordovico-Silurian	43 (6)	42 (1)	34-85
Cambrian	25 (3)	41 (1)	15-41

Table 2. <sup>226</sup>Ra activity in soil (Bq/kg) according to the soil textural class. N=number of data.

Class	ISIB mean (N)	IHE-SCK mean (N)	range
U	-	-	-
E	50 (1)	-	50
A	41 (41)	39 (5)	25-49
L	19 (5)	27 (2)	9-33
P	-	23 (2)	23
S	14 (1)	23 (1)	14-23
Z	10 (5)	12 (6)	6-21
G	39 (15)	37 (9)	22-85

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# The $^{137}\text{Cs}$ repeated sampling approach to derive soil redistribution rates and validate reference sites in alpine grasslands

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**Keywords:**  $^{137}\text{Cs}$  Cesium, soil erosion, Alps, resampling.

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Over the past decades, radioactive  $^{137}\text{Cs}$  fallout has been widely used as a tracer to provide information on soil erosion and sedimentation rates. However, the method may produce relatively large uncertainties in Alpine grasslands. The latter difficulties are caused by a combination of (i) the heterogeneous distribution of atmospheric  $^{137}\text{Cs}$  Chernobyl fallout, (ii) the partly snow covered ground in Alpine areas during the fallout event in April 1986, which results in inhomogeneous  $^{137}\text{Cs}$  distribution during snow melt and (iii) uncertainties in finding undisturbed reference sites in the geomorphological and anthropogenic highly active slopes of the Alps (Alewell *et al.*, 2014).

To overcome these difficulties, in this study we aim to replace the classical  $^{137}\text{Cs}$  approach, where an undisturbed reference site is compared to erosional sites, with a repeated sampling approach (Porto *et al.*, 2014), where we re-sample sites which have already been measured for  $^{137}\text{Cs}$  inventories in the past. Thus, we make use of a temporal instead of a spatial reference.

The repeated sampling approach also allows a quantitative assessment of soil erosion and deposition processes. In this case, the difference between the  $^{137}\text{Cs}$  inventories of a certain point measured in different times is evaluated and employed in a conversion model to derive soil redistribution rates within the study area. In this way information on short term soil redistribution rates is produced.

In addition, the repeated sampling approach can assist the classical  $^{137}\text{Cs}$  approach. For a successful execution of a  $^{137}\text{Cs}$  based erosion study the selection of appropriate reference sites is a crucial step, as a wrong choice may bias the final results. For reference sites it is usually assumed that no soil redistribution processes have occurred since the  $^{137}\text{Cs}$  main deposition.

Therefore, the second aim of this study is to apply the repeated sampling approach to verify the stability of reference sites which can be used for the classical  $^{137}\text{Cs}$  approach and to assess soil redistribution rates since the main  $^{137}\text{Cs}$  fallout.

Two study areas in the Swiss Alps have been investigated: the Urseren valley in Central Switzerland (Canton Uri), with elevation ranging from 1440 to 3200 m a.s.l., and the Piora valley in the southern part of the Alps (Canton Ticino) with elevation ranging from 1850 to 2773 m a.s.l. In Urseren valley six reference sites have been sampled in 2010 and 2013, whereas in Piora Valley four reference sites have been sampled in 2010

and 2014. 9 (Urseren valley) and 12 (Piora valley) sampling sites have been sampled in 2007 and 2014 (Urseren Valley) and 2010-2014 (Piora valley).

Quantitative assessment of soil redistribution resulted in erosion rates ranging from 3.2 to 9.4 t ha<sup>-1</sup> yr<sup>-1</sup> in the Urseren valley. In Piora valley both sedimentation and erosion processes affected the sites in the time window investigated, with mean redistribution rates of 3.2 t ha<sup>-1</sup> yr<sup>-1</sup> (deposition) and 11.4 t ha<sup>-1</sup> yr<sup>-1</sup> (erosion).

The comparison of the reference site inventories measured in different times show significant discrepancy. Particularly high small scale heterogeneity of  $^{137}\text{Cs}$  distribution in the top soil has been found in Piora valley. In the Urseren valley among six sites, only four reference sites are suitable for  $^{137}\text{Cs}$  application following the classical approach.

The  $^{137}\text{Cs}$  repeated sampling approach represents an effective and reliable method to assess short term erosion in Alpine grasslands, and a useful addition for the  $^{137}\text{Cs}$  classical approach, in validating the suitability of reference sites.

## Acknowledgments

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# Conceptual model to categorize and map vulnerability of soils to $^{137}\text{Cs}$ contamination

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**Keywords:** Radiovulnerability, Pantanal, Soil to plant transfer factor, Cs-137, Radioprotection.  
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The complex behaviour of radionuclides in soils makes the prediction of its transfer to plants difficult. Despite this complexity, it is possible to observe a trend on radionuclides behaviour in the soil/plant system (Wasserman et al., 2002; Wasserman, 2009; Frissel et al., 2002). Indeed, radioecological studies have shown that soil properties such as high acidity, very low organic matter content, low fertility and high Fe-Al oxide content, enhances the transfer of  $^{137}\text{Cs}$  to plants (Frissel et al., 2002; Wasserman et al., 2008).

In this work, a conceptual model was established based on literature review concerning soil to plant transfer factor (TF) for  $^{137}\text{Cs}$ . The categories of vulnerability were defined considering variables that are correlated directly or indirectly with the mobility of  $^{137}\text{Cs}$  in the soil/plant system (exchangeable K, cation exchange capacity and pH) and their amplitude of action and interaction on transfer processes (Picanço-Junior, 2011). Based on that, four categories of vulnerability were identified for  $^{137}\text{Cs}$ : 1) Extreme: where TF for cereals are expected to be  $\geq 1$  and remains high even with the soil ageing; 2) High: where TF for cereals are expected to be between  $<1$  and  $\geq 0.1$ ; 3) Moderate: where TF for cereals are expected to be between  $<0.1$  and  $\geq 0.01$ ; 4) Low: where TF for cereals are expected to be  $<0.01$  and reduces with the soil ageing.

The municipality of Jaraguari (MS, Brazil), located at the Pantanal, was selected as the study area. This region is characterized by the diversity of classes of soils typical of humid tropical regions, distributed in the same municipality. The Pantanal ecosystem is recognized as Wetland of International Importance by the Ramsar Convention of the United Nations.

The requirements to apply this conceptual model to classify and to map radiovulnerability were: 1) the ARCGIS platform adapted and automated specifically for  $^{137}\text{Cs}$ , by using logical arguments that defines each category (table 1); 2) a digital map of soil classes for a given area; and 3) a pedological database associate to area.

Table 1: Limits of influence of critical pedological parameter establish for each vulnerability class.

Classes of radiovulnerability	K (cmol/dm <sup>3</sup> )	pH	CTC (cmol/dm <sup>3</sup> )
Extreme FT $\geq 1$	$< 0.05$	$< 12.0$	$\leq 10$
High $0.1 \leq \text{FT} < 1$	$\geq 0.05$ $< 0.30$	$\leq 4.8$	$\leq 10$
Moderate $0.01 \leq \text{FT} < 0.1$	$\geq 0.05$ $< 0.30$	$\leq 4.8$	$> 10$
Low FT $< 0.01$	$\geq 0.30$	$> 4.8$	$> 10$

The Neosols are the dominant soils in the Jaraguari region. It occurs associated to Red Ferralsols, Nitosols, Vertisols and Plinthosol. The conceptual model for classification of soil vulnerability to  $^{137}\text{Cs}$ , based on the pedological parameters, generated a vulnerability map for the Jaraguari County (Figure 1). This map shows that the major part of Jaraguari's region presents low vulnerability to  $^{137}\text{Cs}$  contamination. However few and small vulnerable areas occurring at Jaraguari County could have a key role to spread contamination, since in this region the groundwater table frequently reaches the surface.

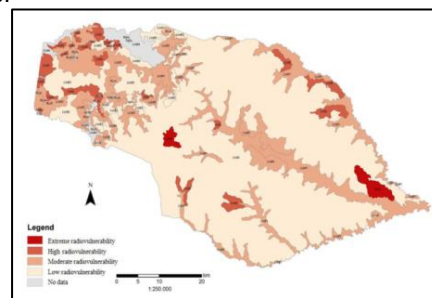


Figure 1: Vulnerability of Jaraguari's soils to  $^{137}\text{Cs}$ .

This result identifies the Pantanal as one of the less vulnerable regions to radioactive contamination, compared with other Brazilian soils, where dominant soils (Ferralsol and Acrisol) were classified as extreme or high vulnerable soil for  $^{137}\text{Cs}$  contamination, since TF for  $^{137}\text{Cs}$  are in general  $\geq 0.1$  (Wasserman, 2009).

This tool showed to be very resourceful for optimizing radiological protection of agricultural areas, once mapping areas of vulnerability and critical soil properties related to  $^{137}\text{Cs}$  behaviour it is possible to establish the geographic regions where remediation action should be prioritized.

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# An analysis of intra-species and inter-species differences in uptake of radioiodine from soil by flowering plants

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**Keywords:** radioiodine, transfer factor, plant uptake.

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Radioiodine has been important in several significant releases of radioactivity to the environment. Both the Chernobyl and Fukushima accidents released significant amounts of <sup>131</sup>I, an isotope with a short half-life (8d) that can, via direct foliar deposition, contaminate plants and food chains. <sup>129</sup>I has a much longer half-life (15.6 x 10<sup>6</sup> y) and is a significant component not just of accidental releases but also of medium/high level nuclear waste. The soil to plant transfer of <sup>129</sup>I is important not just because over time it can reach the soil, but also because it is one of the potentially mobile radioisotopes in nuclear waste and releases of it from waste repositories might reach surface layers. Here we analyse inter-taxa differences in radioiodine uptake because inter-taxa differences have proven to be useful in predicting the transfer of other radioisotopes to plant and animal species (Beresford *et al.*, 2013).

Iodine is generally mobile in the soil plant system, especially under anoxic conditions. It frequently occurs as IO<sub>3</sub><sup>-</sup> but also as I<sup>-</sup> in the soil solution. Plants, although they do not have an essential requirement for I, take up I<sup>-</sup> rapidly, most likely via mechanisms that evolved for the uptake of Cl<sup>-</sup>. Radioiodine can form a suite of compounds with organic components of the soil solution that decrease its availability to plants, but in general it is one of the mobile radioelements that is available to plants. Other mobile radioelements that are also available for plant uptake include <sup>36</sup>Cl, <sup>99</sup>Tc and <sup>79</sup>Se. All these isotopes can be significant components of nuclear waste and predicting their movement in the environment, including the soil-plant system, can be important in establishing safety cases for waste repositories.

Previous work has shown that for many radioisotopes in the soil-plant system there are significant inter-taxa differences in plant uptake (Willey, 2014), these differences occur both above the species level and below the species level (Penrose *et al.*, 2015). In fact, the inter-species and intra-species differences are sufficient to confirm the hypothesis that there is no reason to regard the species as a particularly useful unit when trying to predict the transfer of radioisotopes from soil to plant. The species is most often defined as a reproductive unit, so there is no particular reason why it should have a strong link to differences in radioisotope uptake.

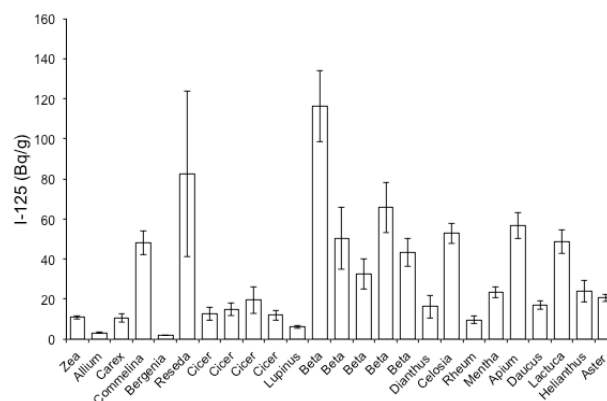


Figure 1. The mean activity of <sup>125</sup>I in 26 taxa of plants following acute exposure through soil under controlled conditions. (n=5, SEs shown, labelled by genera).

We measured, under controlled conditions, the uptake of <sup>125</sup>I by 26 taxa of plants (Fig 1). The species were chosen to represent the major groups of the angiosperm phylogeny and for two of them we measured uptake in 4 and 5 different varieties respectively. We then analysed the inter-species and intra-species variation, and compared differences in iodine uptake with previously published taxonomic analyses of other mobile radioisotopes <sup>99</sup>Tc and <sup>36</sup>Cl. We show that there are significant taxonomic differences in radioiodine uptake and relate these to taxonomic levels above the species and to the behaviour of other mobile radioisotopes.

## Acknowledgments

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# Investigating the role of natural organic matter (NOM) in the migration of key radionuclides in South Terras, Cornwall, England

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The primary focus of this study is on defining the key physical and chemical processes that govern radionuclide transport in the soil solution phase and transfers to biota (Livens & Keith-Roach, 2002, IAEA, 2003, de Boulois et al., 2008).

In this research, the soil quality of the abandoned South Terras mine, Cornwall, SW England (Fig.1) was investigated. Soil core samples up to the depth of 120 cm were collected from 8 different sampling stations and analysed for  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in conjunction with various physicochemical properties of the soils such as NOM and particle content. The radioactivity concentrations in the soil samples were measured using  $\gamma$  - spectrometry for radium and ICP-MS for uranium (Ostle & Cosgrove, 1953).



Figure 1. Cornwall, SW England, UK.

The South Terras mine (Fig. 2) site operated between 1889 and 1930. The mine initially produced uranium ore then, later in its life radium. The site has been abandoned since the mid-1930s and become overgrown, but substantially elevated levels of natural radioactivity can be found in soils (Dean, 1966, Hooker et al., 1989).

One of the steps towards understanding the radionuclide migration through the surface environment is soil characterisation by means of their NOM and soil texture content. The  $^{226}\text{Ra}$  average concentrations widely varied from 0.32 to 6.08 Bq/g, while  $^{228}\text{Ra}$  concentrations varied from 0.041 to 0.059 Bq/g.  $^{238}\text{U}$  average concentrations for the same samples was reported within the range from 1.2 to 49.21 mg kg<sup>-1</sup>.

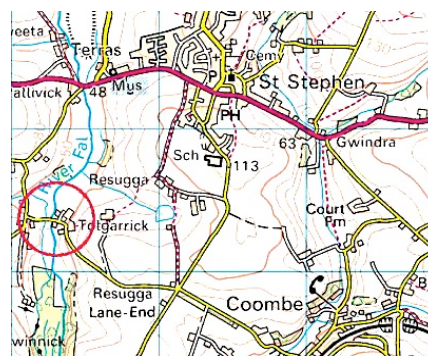


Figure 2. South Terras mine site.

At the same time the NOM values varied from 0.39 to 9.07%. On the basis of the soil texture, the majority of soil samples were characterised as sandy loam and vice versa with sand particles recorded about 80%.

The data analyses indicated that NOM and the above radionuclides correlate in depth variation. Furthermore, the NOM content is slightly increasing in depth while the radium concentrations decrease in depth and the uranium concentrations in most samples shows rapid decrease with depth.

## Acknowledgments

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# Radiocarbon monitoring in the vicinity of the Hungarian NPP

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**Keywords:** Radiocarbon, environmental samples, nuclear power plant.

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The aim of this study was to develop a device, for the radiocarbon determination by LSC by capture of CO<sub>2</sub> from the carbonate content of environmental water samples. Measurements of radiocarbon near NPP-s are a vital part of the environmental monitoring systems.

The developed device is able to release the CO<sub>2</sub> gas trapped in the water in the form of carbonates, then absorb it in a medium (CarboSorb–Perkin Elmer) designed for the absorption of this gas. The absorbed gas was analysed with liquid scintillation spectrometry, the instrument used was a Quantulus 1220.

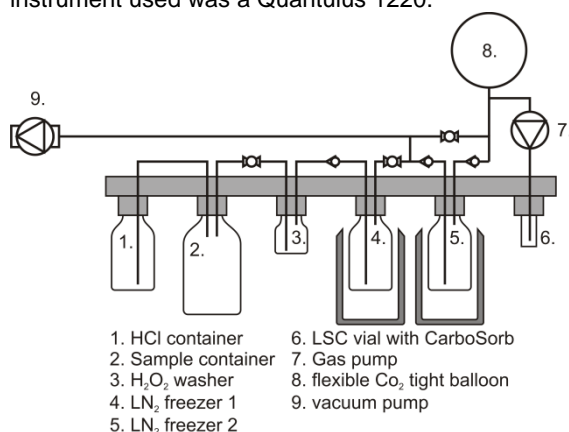


Figure 1. Schematic diagram of the device

The yield of absorption and the all efficiency of the device were determined, than the control measurements of the NPP were commenced. Absorption efficiency was determined with two different methods: the first was to gravimetrically determine the absorbed gas content from the known input carbonate. In this measurement the efficiency was determined from the ratio of the known input and measured output of the system. The second method included the use of a known amount of radiocarbon. This way the efficiency can be determined directly from the measured counts by the LSC.

Sampling were carried out in the vicinity of the NPP, the sampling sites marked on the map, and comparing it with samples wick are taken from the Veszprém region. The water samples were taken from surface waters and the variation of the radiocarbon

concentration in the function of sampling time is essential due to the huge seasonal variation of precipitation.

To determine the variation, the samples were taken from the same places on a fixed period of time, which is set as 3 months. The sampling times are set to be in the middle of meteorological seasons. The measurements are carried out continually, the results presented here include the samples taken at 2015. 01. 15.

As the results show, there is no significant relationship between the radiocarbon activity concentration of the chosen background (Veszprém) region and the area of the Paks NPP. Comparing the results obtained from the same region shows that the deviation of the overall low activity concentration are coming only from statistical reasons.

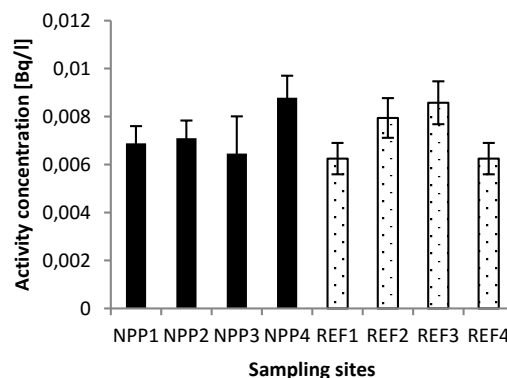


Figure 2. A series of activity concentration measurements comparing radiocarbon activity concentration near the NPP and a reference area

This way we proved that the radiocarbon activity found in the outskirts of the Paks NPP are coming only from natural sources, and are not related to the plant itself.



# Assessment of soil erosion rate based on radionuclide activity concentrations

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Keywords: soil erosion,  $^{137}\text{Cs}$ , gamma spectrometry, river Danube bank.

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In the last decade, research of soil erosion based on measurement of activity concentrations of radionuclides in soil has widely expanded, because this contemporary method is very fast and provides precise data on annual erosion rate. Study of soil erosion is extremely important as it affects agricultural soil degradation, leads to pollution and destruction of water springs and permanent changes in the environment.

Measurement of activity concentration of radionuclides in soil, especially  $^{137}\text{Cs}$ , enables quantitative estimation of soil erosion and average deposit on agricultural land over a long period (30-40 years), without the need for long term geological monitoring. Methods of erosion analysis which combine determination of  $^{137}\text{Cs}$  along with other radionuclides from the environment, such as  $^{210}\text{Pb}$  and  $^7\text{Be}$ , are important in order to obtain spatial and temporal distribution of soil degradation. Such information is of great importance for soil preservation measures, whose main priority is to control and minimize soil erosion, and also for development of strategies for sustainable management of river basins and environmental protection.

Application of gamma spectrometry measurements of  $^{137}\text{Cs}$  activity concentration is based on the fact that this artificial radionuclide has been uniformly spread and adsorbed on surface layer of soil, during the period of nuclear testing (1950-1960) and nuclear accidents (Chernobyl nuclear power plant disaster in 1986). Subsequent redistribution of  $^{137}\text{Cs}$  indicates soil erosion and provides the possibility for precise determination of annual soil deposition.

Having in mind the variety of procedures and models, used for estimation of erosion rate or deposition, based on measured  $^{137}\text{Cs}$  activity concentrations, a distinct difference between agricultural soil and non-agricultural soil (meadow, pasture) should be emphasized. In case of agricultural soil, due to tillage, the mixing of surface and deep soil layers occurs which eliminates the difference in  $^{137}\text{Cs}$  activity concentration among soil layers. On the other hand,  $^{137}\text{Cs}$  is concentrated in surface layer of non-agricultural soil, within 10 cm depth, which reflects the atmospheric artificial origin of this radionuclide. In most non-agricultural soil, total  $^{137}\text{Cs}$  activity per square meter is contained near the surface (within 10 cm depth) and decreases exponentially with depth.

The right bank of river Danube, starting from town of Neštin to Slankamen town, is prone to landslides, which endangers buildings built there and also safety of people living in that area. We have selected two parallel slopes as potential erosion areas, and 4-5 representative points on these slopes for soil sampling. For analysis of soil erosion, we performed sampling in a way to create a depth profile of soil, with 10 cm thick layers, up to 50 cm

depth. The highest sampling point represents a reference point at which is assumed that erosion is not present and that total  $^{137}\text{Cs}$  activity per square meter gives total deposition of this artificial radionuclide for specified area. Other points are equally distributed along the slope, at adequate distance to reveal differences in  $^{137}\text{Cs}$  activity concentration in soil. Soil samples were dried at  $105^\circ\text{C}$ , milled and homogenized. Typical mass of samples was 200g-300g. Activity concentration of radionuclides was determined using low-level gamma spectrometry.

Total  $^{137}\text{Cs}$  activity per square meter for sampling points were calculated as following:

$$^{137}\text{Cs}_{\text{inventory}} = \sum_{i=1}^n C_i \cdot BD_i \cdot D_i$$

i – number of layer, n – maximum number of layers with registered  $^{137}\text{Cs}$  concentration,  $C_i$  –  $^{137}\text{Cs}$  activity concentration (Bq/kg) for particular layer,  $BD_i$  – density of dried soil ( $\text{kg}/\text{m}^3$ ) for particular layer,  $D_i$  – layer thickness (m).

Reduction of total  $^{137}\text{Cs}$  activity per square meter relative to reference value is derived from:

$$X = \frac{A_{\text{ref}} - A}{A_{\text{ref}}} \cdot 100$$

$A_{\text{ref}}$  – total  $^{137}\text{Cs}$  activity per square meter for reference point ( $\text{Bq}/\text{m}^2$ ),  $A$  – total  $^{137}\text{Cs}$  activity per square meter for other sampling points ( $\text{Bq}/\text{m}^2$ ).

Soil erosion rate can be estimated for used model (soil profile distribution model) based on the following equation:

$$Y = \frac{10}{t - 1986} \ln \left( 1 - \frac{X}{100} \right) h_0$$

Y – annual soil loss [ $\text{t}/\text{ha}/\text{yr}$ ], t – year of sampling,  $h_0$  – factor which describes the shape of profile [ $\text{kg}/\text{m}^2$ ].

Estimation of erosion rate for two slopes at river Danube right bank, near the village Ribnjak is presented in table 1.

Table 1. Estimated soil loss for 2 selected slopes.

Slope	Sampling point	$^{137}\text{Cs}$ inventory [ $\text{Bq}/\text{m}^2$ ]	X [%] reduction	Soil loss Y [ $\text{t}/\text{ha}/\text{yr}$ ];
1	1	1392.79	79.8%	1.123
	2	2879.94	58.1%	1.415
	3	2456.78	64.3%	1.351
2	6	2500.83	32.5%	1.564
	7	4972.37	-34.2% (deposition)	1.839 (deposition)
	8	3238.48	12.6%	1.667

# Validation of the method of determination of $^{226}\text{Ra}$ in NORM by LSC: comparison with ICP-MS and gamma spectrometer method

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Keywords: NORM, Ra-226, LSC, gamma spectrometer

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$^{226}\text{Ra}$  analysis is of particular interest for radiation protection, environmental and geological purposes. In worldwide, the radioactivity of  $^{226}\text{Ra}$  in raw materials and by-products has been controlled by national regulators. We developed the  $^{226}\text{Ra}$  determination method by LSC and analyzed  $^{226}\text{Ra}$  in standard reference materials with similar type of raw/by product materials in the industry field. These results were compared with ICP-MS and gamma spectrometer method.

Samples are decomposed by Fusion  $\text{LiBO}_2$  for complete decomposition of mixtures of silicates and zircon (Bojanowski et al, 2002).  $^{226}\text{Ra}$  was extracted from sample by coprecipitation with Ba as sulphate.  $\text{Ba(Ra)SO}_4$  converts to  $\text{Ba(Ra)CO}_3$  with  $\text{K}_2\text{CO}_3$  (Burnett and Tai, 1992) and is dissolved in weak  $\text{HNO}_3$  solution (Fig 1). The source for  $^{226}\text{Ra}$  measurement by LSC is prepared with a water-immiscible cocktail (Maxilight, Hidex).  $^{222}\text{Rn}$  emanation method was applied for determination of  $^{226}\text{Ra}$  using water-immiscible cocktail (Maxilight, Hidex) by LSC (Quantulus 1220).  $^{222}\text{Rn}$  as decay product of  $^{226}\text{Ra}$  is extracted from sample into cocktail, while other radionuclides remain in the sample.  $^{133}\text{Ba}$  is used as a tracer and measured by HPGe detector.

$^{226}\text{Ra}$  is determined through  $^{222}\text{Rn}$  emanation method by LSC using alpha/beta separation. The counting efficiency for  $^{222}\text{Rn}$  and its decay progeny is  $238\% \pm 13\%$  (1 SD) and constant during  $^{222}\text{Rn}$  ingrowth.

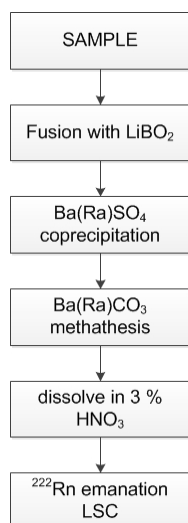


Figure 1. Schematic flow diagram of  $^{226}\text{Ra}$  determination by LSC

We analyzed  $^{226}\text{Ra}$  in CRM 388 (Zircon, Bureau of Analyzed Sampled Ltd.), SRM 1633c (Coal Fly Ash, NIST) and SRM 600 (Bauxite, NIST) to validate this method. Recovery using  $^{133}\text{Ba}$  was  $82.0\% \pm 10.8\%$  (1 standard deviation,  $n=25$ ). The relative error between  $^{226}\text{Ra}$  by LSC and  $^{238}\text{U}$  by ICP-MS is within 10 %.

Table 1. Massic activity of CRM and SRMs

	CRM 388 (BCS)	SRM 1633c (NIST)	SRM 600 (NIST)
Nuclide	Activity ( $\text{Bq kg}^{-1}$ )		
$^{238}\text{U}$	3561.2 <sup>a</sup>	114.4 <sup>a</sup>	128.5 <sup>b</sup>
$^{226}\text{Ra}$	3809.0	114.5	106.9

a:  $^{238}\text{U}$  assigned by U mass ratio (0.00756 from ICP-MS)

b: no information in certificates, assigned by ICP-MS

Gamma spectrometer equipped with HPGe detector was used to determine  $^{226}\text{Ra}$  in same samples by measuring g-ray of  $^{214}\text{Bi}$ . The difference of  $^{226}\text{Ra}$  results between LSC method and HPGe analysis is within 10 % and it shows good correlation (Fig 2).

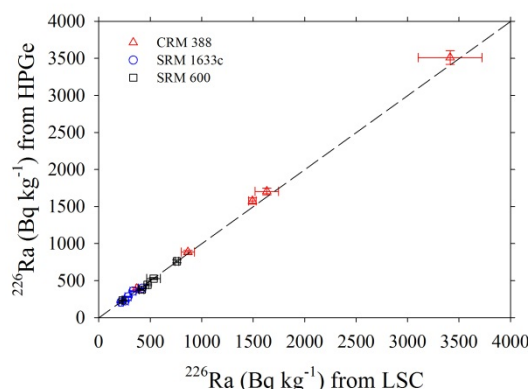


Figure 2. Comparison of  $^{226}\text{Ra}$  results between LSC method and HPGe detector.

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# Observation and modelling of temporal variation in radon decay product concentration in rain water

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**Keywords:** wet deposition, radon decay products, scavenging efficiency, dynamical model.

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Radon-222 and its decay products (hereafter, referred to as RDP) have long been used as tracers of local to inter-regional scale atmospheric transport of contaminants including deposition processes. The RDP concentration in rain can be used to evaluate efficiency of wet deposition. It has been said that the RDP concentration in rain tends to decrease with increasing rain intensity. However, our recent observation showed a positive correlation between them. The purpose of this study is, therefore, to understand and to model the wet deposition processes of RDP, based on continuous measurements of RDP concentrations in rain water.

Measurement was carried out in the Higashiyama Campus of Nagoya University (35.15°N, 136.97°E) during the one year period from December, 2013. The procedure of measurement was repeated every 15 minutes when the rain gauge detected rain, during which 15 mL of rain water underwent gamma spectrometry for 6 minutes with a 2 inch  $\phi \times 2$  inch well-type NaI(Tl) detector, followed by flushing and a 6 minute background measurement. In parallel with this measurement procedure, rain water was collected with a funnel with a diameter of 30 cm to provide a rain water sample for the next round of measurement. Concentrations of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  were evaluated from pulse height distributions.

During the one year period, 37 rain events were observed that had at least four consecutive data points (1 hour duration) with rain intensity higher than  $0.85 \text{ mm h}^{-1}$ . The average  $^{214}\text{Pb}$  plus  $^{214}\text{Bi}$  activity concentration was  $0.22 \text{ Bq mL}^{-1}$  and the average  $^{214}\text{Bi}/^{214}\text{Pb}$  ratio was 1.01. The monthly average of activity concentration did not show significant seasonal variation. Our previous study showed relatively high concentrations in winter reflecting frequent long-range transport of radon from the Asian Continent (Yamazawa, et al., 2004). A plausible reason for this discrepancy is that the rain events observed in warm season in this study were more frequently associated with convective rains, for which our previous study showed the concentration in rain tend to be high.

An example of measured concentration in rain water was shown in Figure 1. The observed concentration roughly follow the temporal change in rain intensity, implying positive correlation between them. This kind of correlation was found in the events with relatively low rain intensity typically around a few  $\text{mm h}^{-1}$ . While, in the cases where rain intensity is relatively high, it was found as a relatively common feature that the concentration increased with increasing rain intensity during the beginning of the rain, but the concentration became uncorrelated with the rain intensity after a certain time period of medium to strong intensity of rain.

A simple model of RDP deposition was applied to the observed cases. This model describes temporal

changes in abundance of RDP in an air column in three phases, namely, the cloud particle phase, the falling rain droplet phase and the ground surface water phase, the last one corresponding to the sample water collected in the funnel and introduced to the detector. Each phase is expressed a set of ordinary differential equations describing the change in abundance of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  decaying in series and transfer between phases. The key parameter of the model is the transfer coefficient from the cloud particle phase to the rain droplet phase, which is not exactly the same with but closely related to the conventional scavenging efficiency. It was determined in this study by trial-and-error to be a power function of rain intensity with the exponent of about 0.7.

An example of the simulation results for weak rain cases is shown in Figure 1. The measured variation in the concentration was reproduced by the model well. Also reproduced by the model was the aforementioned less correlation in the medium to strong rain cases, which was analysed to be caused by depletion of the inventory in the cloud droplet phase.

From the observational results and the discussion using a numerical model presented in this paper, it can be concluded that the RDP concentration in rain is determined not only by the scavenging efficiency, which in turn depends on rain intensity, but also by inventory or availability of RDP in cloud.

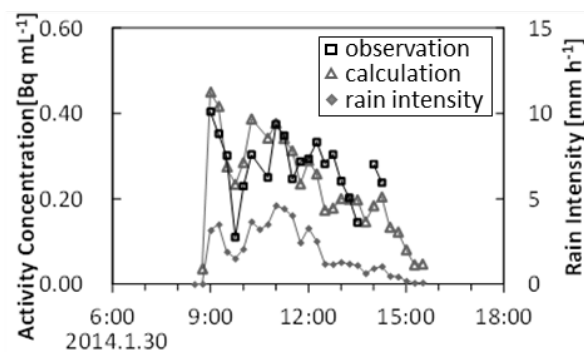


Figure 1. Comparison of temporal change in activity concentration of RDP in rain water between observation and model calculation.

## Acknowledgments

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# A dynamical approach in the distribution patterns of natural radionuclides decaying from Radon-222 in the atmosphere

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Keywords:  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$ ,  $^{210}\text{Po}$ , atmospheric resident time  
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This research introduces a mathematical model of the sources and processes controlling the distribution and transport of the natural radionuclides  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  in the atmosphere.  $^{210}\text{Pb}$  is widely used for dating environmental records stored in natural archives such as lake sediments or peat bogs. In calculating the age of a buried sediment layer, it is first necessary to estimate the initial  $^{210}\text{Pb}$  activity in that layer when it was laid down at the sediment/water interface (Krishnaswami et al., 1971). One key parameter used in making this assessment is the  $^{210}\text{Pb}$  inventory in the sediment core and its relationship to the atmospheric flux (Appleby & Oldfield 1983). Measurements of flux are scarce, and in many parts of the world they are non-existent. The model is validated in the northern hemisphere (specifically between latitudes  $30^\circ\text{N}$  and  $65^\circ\text{N}$ ), where such data is available.

The proposed model is based on a system of four coupled partial differential equations. Since  $^{222}\text{Rn}$  is an inert gas and removed from a column of air only by radioactive decay, its vertical distribution in the column will satisfy the partial differential equation

$$\frac{\partial C_{Rn}}{\partial t} = \frac{\partial}{\partial z} \left( D \frac{\partial C_{Rn}}{\partial z} \right) - \lambda_{Rn} C_{Rn}$$

where  $C_{Rn}(z, t)$  denotes the  $^{222}\text{Rn}$  concentration ( $\text{Bq/m}^3$ ) at altitude  $z$  and time  $t$ ,  $D$  is an effective vertical diffusivity, and  $\lambda_{Rn}$  is the  $^{222}\text{Rn}$  radioactive decay constant. The boundary conditions for this equation are

$$-D \frac{\partial C_{Rn}}{\partial z} \Big|_{(0,t)} = F, \quad C_{Rn}(z, t) \rightarrow 0 \text{ as } z \rightarrow \infty$$

where  $F$  denotes the  $^{222}\text{Rn}$  flux ( $\text{Bq/m}^2$ ) into the base of the column. In contrast to  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$  atoms are highly reactive and readily adsorbed onto dust particles. They may be removed from the atmosphere by wet and dry deposition, as well as by radioactive decay to its daughter radionuclides  $^{210}\text{Bi}$  and  $^{210}\text{Po}$ . The  $^{210}\text{Pb}$  concentration  $C_{Pb}(z, t)$  in the atmosphere is determined by the partial differential equation

$$\frac{\partial C_{Pb}}{\partial t} = \frac{\partial}{\partial z} \left( D \frac{\partial C_{Pb}}{\partial z} \right) + \lambda_{Pb}(C_{Rn} - C_{Pb}) - \Lambda(C_{Pb})$$

and boundary conditions

$$\frac{\partial C_{Pb}}{\partial z} \Big|_{(0,t)} = 0, \quad C_{Pb}(z, t) \rightarrow 0 \text{ as } z \rightarrow \infty$$

where  $\Lambda(C_{Pb})$  is the term characterizing the rate at which  $^{210}\text{Pb}$  condenses from the aerosol state dominated by turbulent diffusion to incipient precipitation dominated by gravity,  $\Lambda(C_{Pb}) = \kappa C_{Pb}(1 - H(z - z_1))$  where  $\kappa$  is the removal rate,  $H(z)$  is the Heaviside function and  $z_1$  is the height of

the tropopause (Piliposian & Appleby 2003). Since the transport processes for  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  can reasonably be presumed to follow those of  $^{210}\text{Pb}$ , the partial equations of  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  will be similar to  $^{210}\text{Pb}$ .

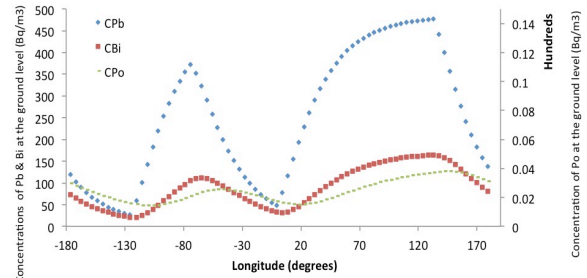


Figure 1. Concentrations of  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  at ground level for 5-day resident time.

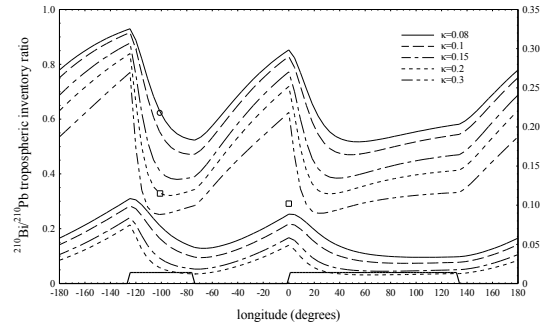


Figure 2. Tropospheric inventory ratios for  $\kappa$  between  $0.08 \text{ d}^{-1}$  to  $0.3 \text{ d}^{-1}$ . The empirical results are shown by the symbols  $\circ$  ( $^{210}\text{Bi}/^{210}\text{Pb}$ ) and  $\square$  ( $^{210}\text{Po}/^{210}\text{Pb}$ ).

The model calculates atmospheric concentrations for long timescales of a year or more and inventories across all longitudes at any given altitude (Fig. 1). Different values of removal rates are examined in order to acquire a reliable resident time through cross-validation with empirical data (Fig. 2).

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# Mapping of the dose rate from ground natural gamma radiation in the Euganean Hills: measured dose and bedrock concentration of radionuclides

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Keywords: Gamma dose rate, natural radionuclides, gamma spectrometry, geostatistic

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An experimental campaign was conducted on the Euganean Hills, near Padua (a city in the Venice area) to assess the dose rate from gamma radiation emitted from the ground, due to the presence of K-40 and of the uranium and thorium decay families (Cinelli et al., 2014).

During the campaign gamma dose rate was measured directly in 103 georeferenced points). The measurements have been conducted with an *Abacus* detector: the latter is comprised of one Halogen-quenched, uncompensated GM pancake tube with thin mica window and one Halogen-quenched, uncompensated thin wall beta-gamma GM peanut tube. The measurements have been taken placing the detector directly next to and in contact with the rock surface.

To estimate the terrestrial gamma dose rate the cosmic contribution to was subtracted, see e.g. Sakellariou 1995.

Using these data a terrestrial gamma dose rate map was developed with geostatistical procedures, and will be presented in this work.

Rock samples were collected at 30 of the 103 locations where the gamma dose rate measurements were taken. The rock samples were subsequently have been analyzed in the laboratory on a HPGe gamma spectrometer to determine radionuclide content. In particular, the thorium series was assessed from its member Ac-228, and the uranium series from Ra-226 - under the assumption that the latter was at equilibrium with the all preceding members of the family (Gilmore, 2008). Uranium, Thorium and K concentration maps have been constructed using geostatistical tools.

The expected dose rate at one meter above the ground for natural emitters can be determined using the conversion factors calculated by Beck (1972) for emitters uniformly distributed in the soil, and reported in the following table 1.

Table 1. Dose rate conversion factors.

Radionuclide	Conversion factor (nSv.h <sup>-1</sup> /mBq.g <sup>-1</sup> )
K-40	0.048
Ra-226 + daughters	0.49
Th-232 + daughters	0.76

For the 30 locations where rock samples were collected the terrestrial gamma dose rate was calculated from the radionuclidic concentrations using the conversion factors above.

The terrestrial gamma dose rate so calculated was compared to the values measured directly.

Both the map and the measured-to-calculated dose comparison will be presented.

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# Distribution of radioactive materials in the sand samples of coastal areas along the Aegean Sea (Greece) and dose assessment of natural radiation

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**Keywords:** Radioactive material, Sand, Environmental radiation, Dose assessment, Aegean sea.

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Humans are being exposed to background radiation from natural and artificial sources daily; however, the natural radiation is the largest contributor to human exposure from the environment. In the present study, the concentration of natural and anthropogenic radionuclide contents in the sand beach samples (Summer 2013) from the Aegean Sea coastal area, located in north-eastern Greece, was evaluated to estimate the effective dose rate from natural radioisotopes and assess the health risk of radiation; meanwhile, the concentration of <sup>137</sup>Cs was measured to discover the influence of the Fukushima Dai-ichi nuclear accident on the man-made isotope concentration level in the Aegean Sea area.

The High-purity Germanium detector (HPGe), with low background, high resolution and high efficiency, was used to detect gamma ray emissions from radionuclides present in the samples. The absolute efficiency of each energy peak was calculated using a mixed radionuclides gamma standard soil (IAEA-375).

The concentrations of <sup>137</sup>Cs and <sup>134</sup>Cs were measured in the range of 0.8±0.2 to 3.3±0.5 Bq/kg and below the MDA (0.3 Bq/kg), respectively; however, the activity concentration of <sup>137</sup>Cs was measured to be lower than the regional background concentration.

The activity concentrations of the main naturally occurring radionuclides of <sup>40</sup>K, <sup>232</sup>Th, <sup>226</sup>Ra and <sup>235</sup>U were determined in the range of 426±20 to 740±28 Bq/kg (565±21 Bq/kg), 9±2 to 67±9 Bq/kg (25±5 Bq/kg), 9±1 to 31±3 Bq/kg (15±2 Bq/kg) and 0.03±0.01 to 0.7±0.1 Bq/kg (0.3±0.1 Bq/kg), respectively.

The total gamma radiation dose was estimated to be in the range of 64±38 to 113±40 nGy/h (81±39 nGy/h), moreover the total gamma absorbed dose rate from naturally occurring radioactive materials was calculated between 29±12 and 77±16 nGy/h with an average of 46±12 nGy/h.

By taking into consideration the activity concentration of radionuclides and the gamma absorbed dose, the radioactivity index, the radium equivalent activity and the annual effective dose were calculated and are shown in Table 1.

Table 1. The radioactivity index, radium equivalent and the annual effective dose in sand samples.

Value	Radioactivity Index	Ra <sub>eq</sub> (Bq/kg)	Annual effective dose (μSv/y)
Min	0.2	59±9	36±14
Max	0.6	161±12	94±20
Mean	0.4	94±10	56±15

In the light of the results, no evidence was determined to prove the impact of radioactive materials leaked from the Fukushima disaster on the Aegean Sea coastal area. The average value of gamma absorbed dose was calculated below the worldwide average value as 55 nGy/h. The radioactivity index for all samples was estimated to be below 1 in accordance with COUNCIL DIRECTIVE 2013/59/EURATOM.

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Et al.

# Radioactivity in special nature reserve “Zasavica”

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**Keywords:** nature reserve, gamma spectrometry method, natural radioactivity, artificial radionuclides.  
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Special nature reserves and protected areas are areas with special natural resources that are most sensitive to changes in the environment caused by human activity. In this paper, the research of natural and artificial radionuclides in the non-agricultural soil, sediment and plant material in special nature reserve Zasavica is presented. In special nature reserve “Zasavica”, stretching from south of Sava river and east of Drina river, at the territory of Sremska Mitrovica municipality, water and swamp ecosystem with fragments of forests and flood-meadows is dominant. With the assistance of expert staff of “Zasavica”, five locations were chosen, where contamination caused by human activities and accumulation/redistribution of radionuclides has been likely. These five locations were selected for the studies of the distribution and possible redistribution of natural radionuclides and artificial radionuclides, such as <sup>137</sup>Cs, which resulted from nuclear accidents in the past.

At each site, dosimetric measurements of ionizing radiation, in-situ gamma spectrometric measurements and sampling of soil, sediment and plant material for gamma-spectrometric low level measurements in the laboratory were performed. In figure 1, selected measurement locations are marked: A - Valjevac – Vizitorski centar, B - Ravnjanska čuprija, C- Raševića čuprija, D - Gajića čuprija, E - Ustava- Mačvanska Mitrovica.

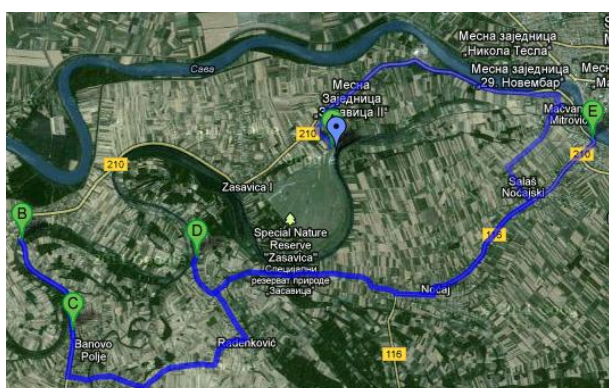


Figure 1. Map with marked measurement locations.

In figure 2 in-situ gamma spectrum at location A is shown. In this spectrum, K-40 line is prominent (at 1460.80 keV), with a few lines from uranium and thorium decay series, characteristic for soil. Recorded in-situ gamma spectra didn't indicate any surface contamination of examined terrain.

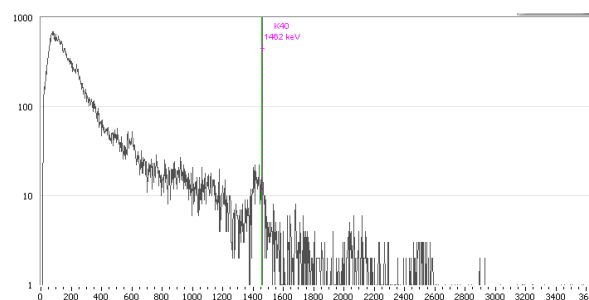


Figure 2. In-situ gamma spectra at location A.

Activity concentration of radionuclides in the non-agricultural soil, sediment and plant material was determined using low-level gamma spectrometry. Samples were dried at 105°C, milled and homogenized. Typical mass of samples was 200g-300g. Each sample was measured by HPGe detectors for 60000 s. Based on gamma line intensities registered in measured spectra, activity concentrations for all radioisotopes that are present in the samples were determined. For radioisotopes that have not been registered by neither of characteristic gamma lines in acquired spectra, upper limit for activity concentration is estimated. Measurement errors are expressed at 95% confidence level.

Determined activity concentrations of natural radionuclides (uranium, radium, thorium, potassium) and artificial radionuclide - cesium in soil and sediment samples are within the usual limits in Vojvodina province. In some samples of plant material elevated levels of NORM radioactivity have been found, due to pre-concentration of radionuclides by increased uptake from sediment. Registered activities of natural radionuclides in soil and sediment are in equilibrium, while artificial radionuclides were not detected or have been detected in traces, implying that there is no radioactive contamination. Measured absorbed dose rates in air are close to natural background rate and, as well as in-situ gamma spectra, prove a fact that examined terrain is not contaminated by natural nor artificial radionuclides. Soil samples from all selected locations do not show elevated levels of radioactivity which would endanger sensitive balance of flora and fauna in this protected area. Activity concentrations of <sup>238</sup>U in surface layer of soil range from 37 Bq/kg to 63 Bq/kg, and are in good agreement with values for soil in Vojvodina province, for which there is a large number of systematic measurements. Elevated activity of <sup>238</sup>U and <sup>235</sup>U, i.e. indication of the presence of depleted uranium is not found.

# Radon concentration measurements in Gattar uranium prospect, Northern Eastern Desert, Egypt

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**Keywords:** radon in mine, Gabal Gattar, Urghada, Solid State Nuclear Track Detector. Radon concentration. Natural radioactive elements.

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In this present work, the radon gas concentration is practically measured in closed uranium prospect mine located at Gabal (G.) Gattar, Salman et.al (1990). CR-39 solid state nuclear track detector technique is used. It is found that the radon concentration is around  $80 \text{ kBqm}^{-3}$  and an effective ventilation rates should be applied if there will be further works in the future, Durrani and Ilic (1987).

The concentration of Radon in mines varies tremendously according to the country rock, type of mineralization and area. Ventilation is also an important factor, since mines with a poor air flow tend to allow a greater concentration of Radon to build up.

The radon gas concentration is practically measured in closed uranium prospect mine located at Gabal (G.) Gattar. It is located some 35 km west of Hurghada city, Red Sea coast. G. Gattar acquired its importance due to hosting numerous uranium occurrences. This work cares about G II, which is localized at the western experimental uranium mine GII occurrence, Shalaby (1995).

To achieve this target, a series of solid state nuclear track detectors (CR-39) were attached to a bottom of stainless steel can (covered by a thin membrane to let only the radon gas enter to the detector) and hanged up to the ceiling of the mine (= 1m from the ground surface) in different places. Two other detectors (A and B) are located outside to determine the background or better the outside radon concentration.

The radon concentration was found to be  $4.81 \text{ kBqm}^{-3}$  and  $10.18 \text{ kBqm}^{-3}$ , respectively. If this mine is on work, the radon gas is controlled by applying effective ventilation. The condition of ventilation is varied each time according to the quantity of dust aiming to reach to the clean atmosphere to protect the miners' health, KIES et.al (1996).

Finally, it can be concluded that the average annual exposure of uranium miners have fallen to level similar to the concentration inhaled in nature.

## Acknowledgments

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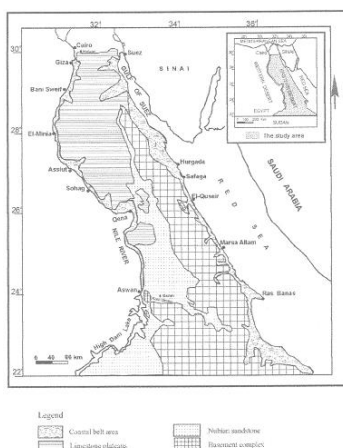


Figure 1: Map of Egypt showing G. Gattar



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## Environmental and radiological risk assessment of the Schistos phosphogypsum waste repository

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**Keywords:** Phosphogypsum, Radiological waste, Risk assessment, GIS.

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The operation of a phosphate fertilizer industry resulted in the deposition of 10 megatons of phosphogypsum (PG) in an abandoned limestone quarry in Schistos, near Piraeus port. The present study aimed at assessing the environmental and radiological impact of the Schistos phosphogypsum (PG), which is known to host elevated amounts of radioactivity. Several analytical and laboratory techniques were applied to the studied PG.

The radioactivity content of PG is mainly due to the <sup>238</sup>U series (<sup>226</sup>Ra in particular), while the majority of the soil samples are found to contain much less radioactivity (mainly to <sup>40</sup>K).

The γ-ray spectrometry results were combined to GPS coordinates to create GIS maps, showing the radionuclides distribution in the studied area and to assess the potential hazard.

The <sup>226</sup>Ra radioactivity content is found to be elevated, mainly in the exposed area, but rather low compared to the global statistics (Tayibi et al., 2009; Papastefanou et al., 2006).

The total absorbed gamma dose rate is found to be 220 nGy/h, exceeding the international recommended and acceptable values (30-109 nGy/h) for workers in the regions containing γ-radiation (UNSCEAR, 2000).

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- UNSCEAR REPORT (2000) Vol. I: *Sources and effects of ionizing radiation*: ANNEX B: Exposures from natural radiation sources.



# Removal of TE-NORM Scales from Gas-Oil-Water Separators in Libyan Oil Fields

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**Keywords:** Cleaning, TE-NORM, Scales, Petrolite TK-14, Petrolite SP-191.

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The paper describes a new method for non-destructive TE-NORM scale removal from gas-oil-water separator plants (GOWSPs) in Libyan oil fields owned and managed by a Libyan oil production company. The procedure being based on more than 10 years experiences (Fawaris, B. H. and Saad, A. A., 2004) is applied as part of TE-NORM management program of that company, particularly in five selected oil fields. The procedure was adopted for cleaning and removal of TE-NORM scales that were formed inside the GOWSP vessels which were in continuous operation for about 15 years without any maintenance action in between.

The cleaning work was performed during a period of one year and a half with three week time intervals for each of the selected GOWSPs. The procedure works in a close cycling mode using a mixture of diesel fuel with two scale inhibitors (Type Petrolite TK-14 and Petrolite SP-191) according to the following steps:

1. Isolate vessel from inlet manifold or wells
2. Flood oil from vessel using treated firewater
3. Drain all water from vessel to the pit
4. Close oil outlet and water outlet valves
5. Start to pump firewater into vessel through sand jet nozzle built-inside the separator
6. After build-up of water, drain valve opened to discharge oil mixed water to the pit and continue to clean the separator until clean water comes out to the pit
7. Isolate gas backpressure system and de-pressurize the vessel
8. Open the top flange and fill with diesel fuel and PetroliteTK-14
9. Tie-in air compressor through sand jet nozzle and agitate the mixture for 24 hours; re-pressurize vessel using the back pressure gas system and empty the separator to the pit
10. Proceed with steps 6, and 7 until water become clean at the outlet

External  $\gamma$  dose rate measurements have been carried out before and after cleaning of five selected GOWSPs. In addition, three solid TE-NORM scale samples were taken from each of the selected GOWSP for  $\gamma$ -spectroscopy analysis in order to assess the concentration of typical TE-NORM isotopes, such as  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ .

The results of  $\gamma$  dose rate measurements are shown in Table 1 for one typical oil field (Al-laheeb). The results for the other oil fields under investigation were similar. Thus, the measurements show the cleaning procedure to be highly effective for reduction of the activity levels of TE-NORM at the various sites under investigation.

Table 1. Mean  $\gamma$  dose rate at various measuring sites on the GOWSP at Al-laheeb oil field before and after cleaning

Measuring site	Mean $\gamma$ dose rate, $\mu\text{Sv/h}$	
	before cleaning	after cleaning
Flow line inlet	0.93	0.40
Separator bottom	54.0	4.45
Oil outlet	0.55	0.15
Water outlet	7.40	1.60

For quantification of cleaning efficiency a cleaning factor CF has been defined (CF = ratio of the net  $\gamma$  dose rate values before and after cleaning). The cleaning factor turned out to be maximal for the separator bottom where the gamma dose rate or the activity deposition is maximal. It is an interesting finding that the cleaning factor for this measuring site is very similar for all oil fields under investigation, the mean of CF being 11.25. On the other hand the cleaning factor is minimal at the flow line inlet with the mean of CF being 4.32. For the two other measuring sites the cleaning factors are ranging between these two mean values.

The  $\gamma$ -spectrometric analysis revealed the presence of significant activities from  $^{226}\text{Ra}$  and its progeny, as well as  $^{228}\text{Ra}$  and its progeny. The concentration levels of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are ranging between 93,6 Bq/g and 257,1 Bq/g and between 14,8 Bq/g and 102,1 Bq/g, respectively, with hard greyish samples showing typically higher activity levels than soft whitish samples.

In summary, the measuring results revealed the cleaning method to be applicable and very successful for removal of TE-NORM scales from all GOWSP components under investigation. Major advantages of this method are (i) removal of TE-NORM scale without any destruction and/or opening of the GOWSP vessels, (ii) minimum contamination of surrounding site, and (iii) negligible external dose rate to the operating team.

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## New Applications in Accelerator Mass Spectrometry using developments of ion source and ion-gas reaction cell technology

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**Keywords:** Accelerator mass spectrometry, ion-gas reactions, ion source, AMS targets.

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The extremely low isotope ratios (as low as  $10^{-16}$ ) achievable by Accelerator Mass Spectrometry (AMS) are largely the result of its ability to remove isobars, both atomic and molecular, of the rare species. In AMS, molecular isobars are destroyed by the charge changing process in the high voltage terminal of the accelerator, but only a few atomic isobars, such as  $^{14}\text{N}(^{14}\text{C})$ ,  $^{26}\text{Mg}(^{26}\text{Al})$  and  $^{129}\text{Xe}(^{129}\text{I})$  can be easily removed – by their inability to form negative ions (anions) in the ion source.

Post-accelerator techniques have been developed to deal with this problem, for example, the use of foils to degrade the energy and change charge in the case of  $^{10}\text{B}(^{10}\text{Be})$  or gas filled magnets for  $^{36}\text{S}(^{36}\text{Cl})$  and other heavier isobar pairs. In the ion source, partial isobar suppression has been achieved with the use of molecular anions containing fluorine as carriers of the analyte element (Zhao et al, 2010). However, a dependence of beam current on ion source design or operating parameters is still under investigation.

Over the past 10 years, attempts to remove the isobars from the pre-accelerator anion beam have shown significant promise. These include the use of lasers to selectively detach the extra electron from the isobar (Forstner et al, 2008) or ion-gas collisions to selectively react with or simply de-ionize the isobar (Eliades et al, 2010). Both techniques require good control of the energy and phase space of the ion beam, so radio-frequency quadrupoles (RFQs) are employed to guide and contain the ion beam and elastic ion-gas collisions in the RFQ field are used to reduce its energy spread.

This presentation will focus on developments using both the molecular fluoride anions and the RFQ gas-ion reaction cell approaches. The proof-of-principle RFQ reaction cell system built at the IsoTrace Laboratory (University of Toronto), known as an Isobar Separator for Anions (ISA) is described in detail by Eliades et al (2010). This has now been moved to the Lalonde Laboratory in Ottawa where it is being incorporated into a second injection line. This line will later be used to implement the pre-commercial ISA built by Isobarex Corp.

The measurement of the isotopes  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$  provide an example of an application for which separation from the abundant isobars of Ba is required. The techniques for reducing the Ba isobars by generating  $\text{CsF}_2^-$  anions in the ion source and for destroying much of the remaining  $\text{BaF}_2^-$  in the ISA are described by Eliades et al (2013). The selection of  $\text{CsF}_2^-$  from the ion source results in a suppression of  $\text{BaF}_2^-$  by  $5 \times 10^{-4}$  and the interaction of these ions with

$\text{NO}_2$  gas at 0.6 Pa in the reaction cell results in a  $\text{BaF}_2^-$  attenuation of  $2 \times 10^{-5}$ .

On a conventional AMS system, the analysis of Cs isotopes is complicated by the use of Cs as the primary beam in the sputter ion source. Even if Rb were substituted for Cs as the sputtering ion, mass analysis of the sputtering beam would be needed to eliminate the introduction of variable quantities of Cs from impurities in the Rb. The closely coupled design of the high current AMS sputter sources makes such analysis challenging. MacDonald et al (2015) have reported an alternate strategy in which a known quantity of an isotope tracer,  $^{134}\text{Cs}$  in this case, is added to the target matrix. As the  $^{134}\text{Cs}$  is a gamma emitter, it can be used to monitor yield throughout the sample preparation processes; on the AMS system, it can be analyzed as a third, slightly less rare isotope in the gas ionization detector to provide a reference for determining the concentration of the  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$  isotopes in the sample.

These measurements were challenged by the low and variable  $\text{CsF}_2^-$  current available from the fluoride target matrices used and a large memory effect which appeared to be connected with a high initial spike in current. Recent searches over a range of  $\text{PbF}_2$  based target matrices and more appropriate ion source operating parameters have resulted in a significant improvement in the stability of the  $\text{CsF}_2^-$  current and an increase of over two orders of magnitude in overall target yield. Details of these developments will be reported in this presentation

### Acknowledgments

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# High precision $^{14}\text{C}$ analyses by small AMS - applications in nuclear environmental protection

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**Keywords:** radiocarbon, nuclear, environmental protection, AMS.

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Radiocarbon ( $^{14}\text{C}$ ) dating is widely applied in environmental protection, geology, hydrology, and climatology. In 2011 a more developed and modern technology took over our old  $^{14}\text{C}$  measuring methods in Hungary, which is based on isotope separation by accelerator mass spectrometry and not on activity measurements. The advantage of the new method is that it requires much less sample quantity (0.1-100 mg) than the radiometric ones. Besides, accelerator mass spectrometry (AMS) technique could provide ten times higher throughput instead of the formerly used methods. A MICADAS type AMS technique dedicated for radiocarbon studies was developed by ETHZ (Zürich, Switzerland) in connection with environmental research, in particular equipped with a gas inlet system to handle  $\text{CO}_2$  sample material. Installation and successful Final Acceptance of the EnvironMICADAS was completed at Debrecen, Hungary were done by the end of 2011 (Molnár *et al*, 2013a). Background is about 46 kyrs BP. Relative uncertainty after a half day measurement of primary standard (Ox-II, NIST 4990C) was better than  $\pm 0.3\%$ . Results of a secondary standard (Oxa-I, NIST 4990) and standard reference material (IAEA-C7 and – C8) met perfectly with the expected values (Table 1.). In the first three year of operation more than 8000 samples were analyzed in the new AMS lab of Hungary.

Table 1. Acceptance Test of the EnvironMICADAS.

Investigated parameters	Results
Extracted Ion Current ( $^{12}\text{C}$ )	$44.2 \pm 7.8 \text{ uA}$
Ion transmission ( $^{12}\text{C}/^{12}\text{C}_\text{L}$ )	$37.9 \pm 0.9 \%$
Oxa-II (NIST SRM 4990C)	$-2355 \pm 17.9 \text{ yrs BP}$
Oxa-I NIST standard (-313 yrs)	$-314.5 \pm 26.2 \text{ yrs BP}$
IAEA-C7 (49.53 pMC)	$49.43 \pm 0.11 \text{ pMC}$
IAEA-C8 (15.03 pMC)	$15.17 \pm 0.05 \text{ pMC}$
Blank samples	$46,300 \pm 1300 \text{ yrs BP}$

A new method for carbonate sample preparation was implemented using He carrier gas and headspace approach. This new way of carbonate preparation was successfully implemented in the automated graphitization equipment of ETHZ (AGE) and also applied like an excellent extension of the MICADAS gas ion source interface (Figure 1). Background was better than ever before, sample preparation is very simple, safe, clean, quick and cost effective. The new style of carbonate preparation was successfully adapted for dissolved carbonate measurements of groundwater

samples and for other liquid solutions containing dissolved carbon (Wacker *et al*, 2013).

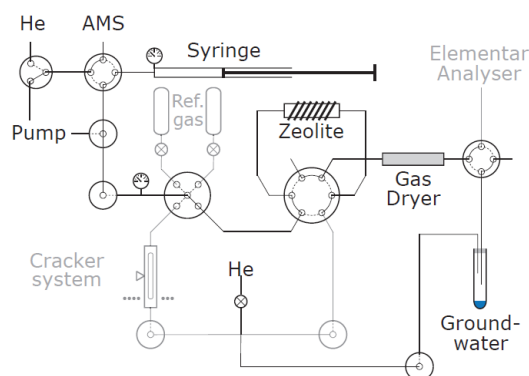


Figure 1. Layout of the connection of ground-water preparation line to the AMS gas ion source interface.

For more than 20 years the radiocarbon and tritium content of atmospheric  $\text{CO}_2$ , water vapour, hydrocarbons and hydrogen have been continuously monitored around the nuclear power plant of Paks (NPP), Hungary. Our laboratory developed the differential radiocarbon and tritium samplers that take integrated samples from the ambient air, there. An automatic groundwater sampling device also has been developed in order to separately take anions and cations by means of ion exchange resins. Having eluted the ions from the resins, cations are measured by gamma-spectrometry in an ultra low background gamma spectrometry laboratory.

Radiocarbon impact in the annual tree rings of a nearby pine tree from 2000-2009 was also studied there and compared to the parallel atmospheric  $^{14}\text{C}$  monitoring observations.

We investigated the impact of low and intermediate level radioactive waste (L/ILW) disposal on the groundwater and ambient air in case of a near filed storage facility (Püspökszilág, Hungary). Thousands of groundwater and air samples for  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{90}\text{Sr}$  and water chemistry have been measured. This work is essential for a safe operation of the radioactive waste disposal facility.

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## Mass spectrometry for Determination of Environmental Radionuclides

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Radionuclides are conventionally measured by detecting their characteristic radiation using alpha spectrometry, beta counting including liquid scintillation counting and gamma spectrometry depending on their decay modes. Accordingly these methods are high sensitive for short half-lived radionuclides. Mass spectrometry, typically used to measure isotopes of elements, can be also used for measurement of radionuclides. In these methods, the atoms of the radionuclide of interest are directly measured. Therefore, mass spectrometry methods are normally sensitive for the measurement of long-lived radionuclides. Among various inorganic mass spectrometric methods, inductively coupled plasma mass spectrometry (ICP-MS) and accelerator mass spectrometry (AMS) are two most popular used mass spectrometry techniques for the measurement of radionuclides, especially long-lived radionuclides. With the improvement of ICP-MS

technique and more instruments to be installed, the application of this technique is becoming more popular tool for measurement of radionuclides. By hyphenation with automated separation system, ICP-MS will play a critical role in rapid determination of radionuclide for emergency analysis. AMS is the most sensitive analytical technique for many long-lived radionuclides, the new development of this techniques, especially the miniaturization of AMS system significantly reduce the cost of instrument as well as maintenance and operation, this stimulated and enhanced the application of this technique in the environmental researches. This work aims to discuss the application of ICP-MS and AMS in the measurement of some most important radionuclides, such as  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{135}\text{Cs}$ ,  $^{236}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , especially the new progress in the analytical methods of these radionuclides for environmental researches.

# The dissolved uranium concentration and $^{234}\text{U}/^{238}\text{U}$ activity ratio in mineral waters of southeastern Brazil

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Keywords: uranium isotopes, U-238 and U-234, mineral waters, southern Brazil.  
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Naturally-occurring uranium isotopes  $^{238}\text{U}$  and  $^{234}\text{U}$  are in secular equilibrium in minerals and rocks older than 1 Ma if the systems are closed for U, implying that the  $^{234}\text{U}/^{238}\text{U}$  activity ratio (AR) is unity in them. However, water-rock/soil interactions frequently result in ARs values above 1 for dissolved U (Osmond and Cowart, 1976).

Oxidizing conditions generally prevail in the recharge zone of an aquifer and the most active U leaching occurs in this zone, where it exhibits a great tendency to oxidize to  $\text{U}^{6+}$  that is very soluble. A groundwater dissolving U close to recharge may undergo further change in its U chemistry due to additional leaching or, under more reducing conditions, due to U precipitation (Osmond and Cowart, 1976; Krauskopf and Bird, 1995). In general, the size, climate, lithology, stratigraphy, hydrogeology, geochemical conditions, and extent of water-rock/soil interactions are among the factors responsible for the variability in the dissolved U content and ARs > 1 values in the aquifer systems (Osmond and Cowart, 1976).

This study reports data of the dissolved U content and AR in 75 mineral water sources occurring in 14 municipalities at São Paulo and Minas Gerais states, southern Brazil, which are extensively used for drinking in public places, bottling and bathing purposes, among other. The results obtained are shown in Table 1, and the most of the values indicated ARs above 1 that may be caused by preferential chemical dissolution of  $^{234}\text{U}$  (Rosholt *et al.*, 1996) and/or alpha-recoil release of  $^{234}\text{Th}$  at the rock/soil-water interface (Kigoshi, 1971), among other factors.

Table 1. The dissolved U content and  $^{234}\text{U}/^{238}\text{U}$  activity ratio in the groundwaters analysed.

Value	$^{238}\text{U}$ content <sup>1</sup> ( $\mu\text{g.L}^{-1}$ )	$^{234}\text{U}/^{238}\text{U}$ activity ratio <sup>1</sup>
Minimum	0.001	0.98
Maximum	4.82	11.68
Mean	0.17	2.56

<sup>1</sup>Analytical uncertainty  $\pm 10\text{--}15\%$  corresponding to 1 $\sigma$  standard deviation

The mineralization of the studied waters is greatly variable (DR-Dry Residue=11-2898  $\text{mg.L}^{-1}$ ). The ground waters are reducing in character, as shown in Fig. 1. Despite this aspect, active U dissolution has been identified in the various aquifer systems investigated. The highest  $^{238}\text{U}$  activity concentration was 0.06  $\text{Bq.L}^{-1}$  that is ~170 times lower than the guideline reference

value of 10  $\text{Bq.L}^{-1}$  in drinking water as established in 2011 by the WHO. The dissolved U and Ca correlated significantly (Pearson correlation coefficient  $r=0.78$ ), whereas AR correlated significantly with DR ( $r = 0.56$ ). The discharge and DR data of the water sources allowed estimate dissolution rates corresponding to 3.9  $\text{ton.yr}^{-1}$  (AR=0.98) and 145.4  $\text{ton.yr}^{-1}$  (AR=11.68). Thus, higher dissolution rates tended to increase the ARs values in the aquifers. The reciprocal of the dissolved U content (S) and the AR of dissolved uranium allowed to determine mixing volumes of different groundwater masses. Two or three water sources, characterized by their different AR and S values, have produced resulting mixtures fitting straight lines or plotting within triangles formed by the end-members. These results showed the applicability of the natural U-isotopes for mixing calculations in the aquifers studied.

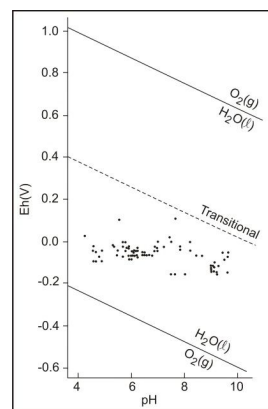


Figure 1. Data of the groundwaters studied plotted in an Eh-pH diagram from Krauskopf and Bird (1995).

## Acknowledgments

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## Natural radionuclides in thermal mineral springs in Edipsos Greece

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Thermal and mineral natural therapeutic springs are appearing at 850 places all over Greece. About 150 of them are active and built spa facilities in different places. One of the most popular thermal spring areas is the city of Loutra Edipsou, located in the northern part of the Island of Evia, known since ancient times for its hot springs. There are more than 80 hot water springs with temperatures varying from 28°C to 86°C. The city of Loutra Edipsou is one of the most visited spa centres in Greece attracting as many as 25000 visitors yearly, at the peak of the tourist season.

These springs, rich in valuable minerals and trace elements, are effective in curing problems such as rheumatoid and inflammatory arthritis, degenerative arthritis, spondylo-arthritis, myalgia, neuralgia, lumbago, neuritis, backaches, tendonitis, vessel diseases, diseases of the endocrine cycle etc.

The thermal and mineral spring waters contain naturally occurring radioactive substances allegedly having beneficial health effects (Falkenbach 2001). However, ingesting or inhaling these radioactive substances may cause serious health issues (Rowland 1993).

On this account a radiological survey has been carried out in the municipality of Edipsos in the year 2004. Considering the strong radiotoxicity of radium-226 and the uranium isotopes the survey has been focused on the identification of those  $\alpha$ -emitters.

Radiochemical results ( $^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{226}\text{Ra}$  activities;  $^{234}\text{U}/^{238}\text{U}$  activity ratio) are reported for 13 thermal spring water samples and 4 tap water samples, collected from 5 locations in the municipality of Edipsos. The obtained results show that  $^{238}\text{U}$  activity varies between 2.23 and 65 mBq/L in hot springs and between 2.75 and 10.58 mBq/L in tap waters. The  $^{234}\text{U}/^{238}\text{U}$  activity ratio lies in the range 1.16 - 2.10 in all analysed water samples. The  $^{226}\text{Ra}$  activity concentration varies between 3.54 and 1470 mBq/L in hot springs and between 4.20 and 16.70 mBq/L in tap waters. The

activity concentration of  $^{226}\text{Ra}$  and the uranium isotopes has been determined using radiochemical separation procedures and measured by alpha spectrometry, Eaton *et al* (1995) and Hancock *et al* (1991).

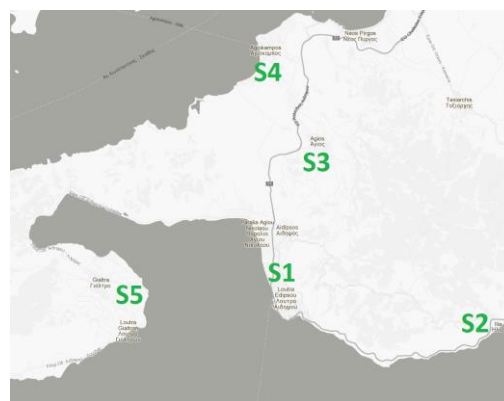


Figure1: Sampling locations in the municipality of Edipsos

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## Character and levels of radioactive contamination of underground water at the Semipalatinsk Test Site

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470 nuclear tests have been conducted at the territory of Semipalatinsk Test Site (STS) including 26 surface, 90 air and 354 underground nuclear explosions (UNE). Underground nuclear explosions have been conducted in horizontal underground workings called tunnels at the «Degelen» site and in vertical underground workings called boreholes at the «Balapan» site.

As a result of nuclear tests conducted there is a huge amount of radioactive products concentrated in blocks of geological environment, containing UNE central zones at the small area. Resulted fields of radioactive contamination significantly change with time, first of all, as a result of different types of radionuclide migration. The main radionuclides, currently participating in the STS underground water contamination are: <sup>3</sup>H, <sup>90</sup>Sr, <sup>137</sup>Cs and <sup>239+240</sup>Pu. The most important and interesting is study of character of radionuclides' migration with underground water, because this process can result in radioactive contamination of potable water sources and loss of geological environment with all its resources.

**«Degelen» site.** Radioactive contamination of the Degelen massif underground water happens due to «wash out» of artificial radionuclides from rocks with atmospheric fallouts, coming to zones of nonreversible deformations through artificial fracture and fault systems. After this, a part of contaminated water comes to the day surface through the tunnel cavity and with spring water, the rest part comes to the ground water basin.

Central cavities of nuclear explosions, conducted in tunnels, are mainly located above the underground water level.

In tunnel waters of «Degelen» site high concentrations of artificial radionuclides have been found. These concentrations significantly overcome permissible levels for drinking water (<sup>137</sup>Cs – up to 700 Bq/l, <sup>90</sup>Sr – up to 2000 Bq/l, <sup>3</sup>H – up to 1300 kBq/l and <sup>239+240</sup>Pu up to 110 Bq/l). Herewith, the common feature is relative stability of these values in time.

Contaminated tunnel waters discharge into natural drain canal – valleys of creeks. These creeks have blind drainages, those limit distribution of artificial radionuclides together with surface and underground waters. All the underground water front, going beyond Degelen mountains, gets divided into individual streams according to their belonging to local watersheds, where feed, transition and discharge appear within the area of basins, with the length up to 20 km.

At the territory, where underground water streams go beyond «Degelen» massif, concentrations of <sup>90</sup>Sr and <sup>137</sup>Cs in water are at the extremely low level.

Fact of absence of high concentrations of <sup>90</sup>Sr and <sup>137</sup>Cs gives an evidence of high sorption capacity of soft sediments filling up river valleys of «Degelen» massif. At the same time, values of <sup>3</sup>H concentrations varies depending on direction. Maximal values on tritium, within the borders of massif, reach 260 kBq/l. At the distance of 10 km from mountains, the concentration of tritium can reach values of 10 - 30 kBq/l, and at the distance of 15 km, the concentration dips down to 0.10 kBq/l.

So, the process of underground water contamination beyond the «Degelen» site is still in progress, and according to radiation monitoring data, it has quite a stable character.

**«Balapan» site.** Unlike UNEs in horizontal tunnels, cavities formed after nuclear blasts conducted in boreholes, are located significantly below the level of underground waters. High temperature in the cavity remains for a long time due to existence of the layer of overlying rocks. Presence of high temperatures facilitates formation of thermal convection. When reaching the cavity, waters get heated, they wash out chemical elements and radionuclides then turn back with them to the aquifer.

Main transporters of UNE radioactive products at the «Balapan» test site are fracture and fracture-vein water. High concentrations of <sup>137</sup>Cs and <sup>90</sup>Sr in underground water have been detected only in immediate vicinity to «warfare» holes. Concentration of <sup>3</sup>H in underground water samples changes from minimal detectable activity (MDA) to values, more than 500 times exceeding permissible level for concentration of this radionuclide in drinking water.

In the streams of underground water running beyond «Balapan» site, concentration of tritium does not exceed 1.0 kBq/l (concentration of <sup>137</sup>Cs and <sup>90</sup>Sr does not exceed MDA). The exception is the area of underground water discharge into the water of Shagan river, where concentration of <sup>3</sup>H reaches 700 kBq/l, that nearly 100 times exceeds permissible levels for drinking water.

At the 10 km section of the riverbed from the border of the STS concentration of tritium gets down step by step to the safe level. The main discharge of radioactively contaminated waters from «Balapan» site into surface waters occurs along the zones of tectonic fault at the territory of feathering-out of the confining stratum – neogene clays. Here we can expect for possible significant changes in concentrations of tritium in the discharge zone, because this migration process depends on hydrogeological factors and amount of atmospheric fallout.



# Tritium activity levels in natural waters around Hannover, Northern Germany

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**Keywords:** tritium, liquid scintillation counting, natural waters, environmental monitoring, Hannover (Germany).  
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Tritium (T) or  $^3\text{H}$ , a radioactive isotope of hydrogen is a pure, low-energy beta emitter ( $E_{\text{max}} = 18.6$  KeV) with a physical half-life of 12.33 y. It is naturally produced by the interaction of cosmic radiation with atmospheric gases (cosmogenic origin), especially from the reaction  $^{14}\text{N}(n, ^3\text{H})^{12}\text{C}$ . However, it can also be produced by man-made activities. For instance during the period of testing of thermonuclear weapons (1950s – 1960s), tritium environmental levels were drastically increased. But after the ban treaty of the atmospheric nuclear tests in 1963, the worldwide tritium levels have dropped at a rate approximately equivalent to its half-life (Cook *et al.* 2003). Nowadays, the on-going nuclear applications (e.g. pressurized water reactors, reprocessing of spent nuclear fuel, etc) also produce and release tritium into the environment. An example is the continuous discharges from the reprocessing facilities in Western Europe, *namely*, La Hague and Sellafield.

Most of tritium spreads in the environment as tritiated water (HTO) and moves within the hydrological cycle. Therefore, environmental monitoring and hydrological studies are the main applications of environmental tritium analysis. In this framework, we are aiming to assess vulnerability of drinking water reservoirs with regard to tritium and its levels as a baseline data for environmental monitoring, i.e. indicator for other contaminants such as radio-iodine.

For this purpose different water samples (surface-, groundwater, and precipitation) were taken from Fuhrberger Feld, a drinking water abstraction area, close to Hannover, Northern Germany (Figure 1). Fuhrberger Feld represents a non-covered water aquifer, which is recharged mainly by rain water. Moreover, it is the main water supply for Hannover city.



Figure 1. Study area: Fuhrberger Feld drinking water abstraction area (30 km northwest of Hannover).

The analysis of tritium in water samples under investigation was based on a liquid scintillation counting (LSC) in a Quantulus 1220. Prior to LSC measurement the samples were distilled after alkaline permanganate treatment to prevent quenching. The counting efficiency

was determined by applying the external standard method using a quench indicating parameter, *viz*, SQP(E) via calibration curves. All water samples were measured for 1200 min to achieve the best counting statistics. The minimum detectable activity (MDA) was calculated as 1 Bq/L for 1200 min.

The mean tritium activity concentrations of the investigated waters are presented in Table 1. All values for tritium levels in Fuhrberger Feld water are substantially below the normative level that is set as a monitoring value for drinking water by the European Union Water Directive (European Union, 1998).

Table 1. Average tritium activity in different waters from Fuhrberger Feld during the period (2013 – 2015).

Type of water	Activity concentration of tritium	
	(Bq/L)	TU
Surface water	1.31	11.10
Groundwater	3.26	27.63
Precipitation	1.60	13.56

Surface water showed tritium concentrations close to the analytical detection limit of our LSC, but still comparable to its levels in rain water. However, they are still higher than the cosmogenic background levels (0.1 to 0.6 Bq/L). This difference can be explained by the influence of fallout from the previous atmospheric nuclear testing. Furthermore, it might be due to the emission from the reprocessing plants in Western Europe. On the other hand tritium in the investigated groundwater revealed higher levels which clearly indicate some bomb tritium.

The very low tritium concentrations in the investigated waters raise no radiological concerns against using the Fuhrberger Feld as a drinking water supply. However the generated data are more relevant for radio-ecological studies, age dating, and monitoring groundwater radio-contaminants such as iodine-129 and chlorine-36.

## Acknowledgments

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# Assessment of character of tritium distribution in air of nuclear testing sites

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Keywords: tritium, air, distribution, contamination, nuclear test site.

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As a result of nuclear tests conducted at the territory of Semipalatinsk Test Site (STS) a huge amount of tritium was formed. In view of the fact that types of explosions and conditions of them perform were different, the amount of produced tritium, routes of entry and character of distribution in the air can significantly differ in each individual case. In previously researches the main sources of tritium entry into the atmosphere were studied in details (water, vegetation, soil moisture, tunnel and well cavities) (Lyakhova et al, 2012).

The main aim of this research was an assessment of contamination of air with tritium and character of tritium distribution in air at the places where various nuclear tests were conducted.

Research works were performed in places of underground nuclear tests (UNE) – «Degelen» and «Balapan» sites, within the zone of radioactively contaminated streamflows – tunnel streamflows and brooks at the «Degelen» site and Shagan river, with high concentration of tritium in water, as well as in places of atmospheric, surface and excavation nuclear tests – «Experimental Field» site and «Atomic» lake.

According to obtained data the most contaminated sites are the zones of radioactively contaminated streamflows.

Level of tritium concentration in the air of a costal zone of tunnel brooks is about  $1000 \div 1300 \text{ Bq/m}^3$ . Significant content of tritium in air is also observed beyond the test site along the riverbeds at the distance of up to 10 km.

Concentration of tritium in the channel of Shagan river changes within wide range from 0.5 to  $500 \text{ Bq/m}^3$ , but with distance from the channel volumetric activity of tritium significantly decreases, achieving background levels.

The area of tritium contamination at the UNE sites is limited by the near-mouth-area of wells and tunnels, with distribution at the distance not over than 500 m from their wellhead. Average volumetric activity of tritium near tunnels does not exceed  $10 \text{ Bq/m}^3$ , but in the wellhead area single values of tritium concentration in air up to  $300 \text{ Bq/m}^3$  were fixed.

The maximal tritium concentration was found inside the body of sealed tunnels. The

concentration of tritium has exponential dependence on the distance. As the distance from the tunnels portal increases the tritium concentration in the air gets higher and maximum level of tritium is determined the close to area of explosion and reaches about  $30000 \text{ Bq/m}^3$  (Figure 1). Exit tritium from the portal of the tunnels is tens of  $\text{Bq/m}^3$  which shows us that tritium can be released from the tunnels' body via rock formations (Lyakhova et al, 2013).

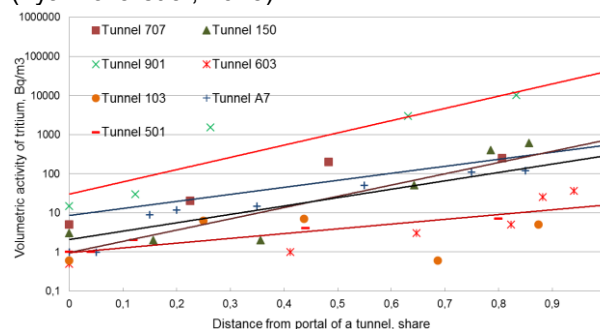


Figure 1. Tritium concentration in the air inside the tunnels body

During research at the «Experimental Field» no significant concentrations of tritium was found in the air. Volumetric activity of HTO did not exceed  $0.5 \text{ Bq/m}^3$ .

At the place of excavation explosion at the territory of «Atomic» lake presence of tritium was also found in the air, volumetric activity of tritium at the cone pile was from 1 to  $90 \text{ Bq/m}^3$ . Distribution of tritium in air at this point has a local character ant limited by the zone of explosion crater location. The research allowed to obtain the entire picture of air contamination with tritium, and also to determine the character of tritium redistribution in air at different radiation hazardous objects of the STS.

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# Natural radionuclides in surface water in a Naturally Occurring Radioactive Material area in Brazil

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In the municipality of Santa Quitéria, Ceará, Brazil, there is a region rich in phosphate associated with uranium and thorium, characterized as a region with Naturally Occurring Radioactive Material - NORM (Pereira, Kelecom & Pereira, 2014).

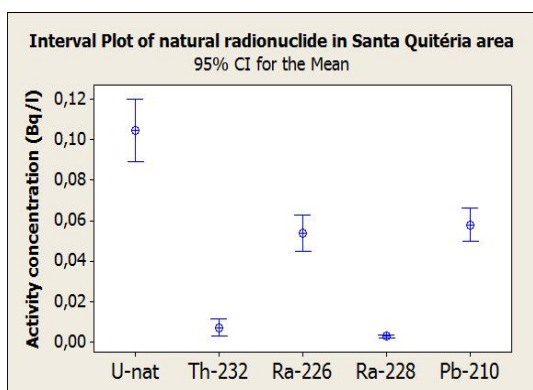
In this paper we analyzed the natural radionuclides most important for committed effective dose to the population in the area of influence of the deposit, namely:  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  (the uranium series), and  $^{232}\text{Th}$  and  $^{228}\text{Ra}$  (thorium series).

A total of 149 samples (per radionuclide) of surface water were collected and analyzed between October 2009 and July 2011 in six surrounding points of the phosphate deposit. The results were assessed by univariate (Ceteno, 1999) and multivariate analyses (Valentin, 2000).

The statistical summary appears in Table 1. The uni-variate analysis evidenced the highest concentrations of activity of the elements from the uranium series when compared to the thorium one. This fact can also be observed in Figure 1.

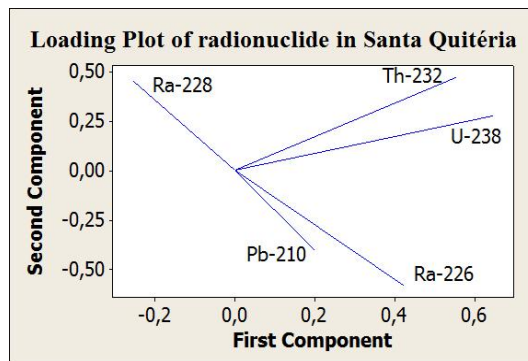
**Table 1** – Statistical summary of natural radionuclides in the NORM deposit area of Santa Quitéria.

Radionuclides	Mean(Bq/l)	Std. Dev.	N
U-nat	0.105	0.096	149
Th-232	0.007	0.026	149
Ra-226	0.054	0.055	149
Ra-228	0.003	0.005	149
Pb-210	0.058	0.051	149



**Figure 1** – Mean concentrations of activity of the natural radionuclides in surface water in the Santa Quitéria region.

PCA (Figure 2) identified three groups: the first one is formed by  $^{238}\text{U}$  and  $^{232}\text{Th}$ , the second by  $^{210}\text{Pb}$ , and  $^{226}\text{Ra}$ , and the third one by  $^{228}\text{Ra}$ .



**Figure 2** – PCA of radionuclides in surface water in the Santa Quitéria region.

All these observations were corroborated by cluster analysis.

The groups formed differed from environmental behavior known for these radionuclides (Lauria, 2007; Klement, 1982).

Comparing the CA of  $^{238}\text{U}$  in waters of this study with the Brazilian means, our values are on the edge of the variation that is of 1-100  $\text{mBq}\cdot\text{l}^{-1}$  (UNESCEAR, 2000). For  $^{226}\text{Ra}$ , our values ( $0.037 \text{ Bq}\cdot\text{l}^{-1}$ ) are 50% higher than world averages (Klement, 1982).

We can conclude that the radioecological behavior of natural radionuclides in this region is complex, and differs from that found in the literature, needing further studies.

The authors are grateful to Fundação Nacional de Desenvolvimento do Ensino Superior Particular - Funadesp at the research fellowships awarded to Prof. Dr. Wagner de Souza Pereira

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# Long-term Radiological Characterization of Waters at one Catalan Drinking Water Treatment Plant

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**Keywords:** water treatment, radioactivity removal, gross alpha, gross beta.

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Radiological characterization of both raw and finished waters at the Sant Joan Despí Drinking Water Treatment Plant (DWTP) has been carried out by the Radiochemical and Radioactive Analysis Laboratory at the Universitat Politècnica de Catalunya since 1993. The DWTP takes waters from the Llobregat River and the Cornellà aquifer is often also used as a complementary source. Sant Feliu aquifer is used for blending at the Llobregat area network.

Measurements were carried out in order to evaluate removal of gross alpha and beta activities by the treatment. In addition, the temporal evolution of gross alpha and gross beta activities is also presented.

The DWTP was built in 1955 and it has been treating water from the lower course of the Llobregat River since it was constructed. Currently, it has a maximum treatment capacity of 5.5 m<sup>3</sup>/s, and provides almost 40% of the annual drinking water in the Barcelona Metropolitan Area. Until 2009 it had been using conventional treatment which essentially consisted on peroxidation, clarification, ozonation, carbon filtration and final chlorination. In 2009 membrane technology using reverse osmosis (RO) was also introduced and since then up to 50 % of the water is treated by membrane technology.

Gross alpha and beta activities in waters were determined by using the methodology detailed in Montaña et al., (2013). All these procedures have been accredited by the Spanish Nacional Accreditation Authority (ENAC).

Figure 1 shows the temporal evolution of gross alpha activity in the Llobregat River and in treated waters in the period 1993-2013. As regards conventional treatment, comparing values between waters from the Llobregat River and finished water, it is difficult to note a decrease in radioactivity in the period 1993-2009 and therefore it can be stated that the classical treatment without membranes is not suitable for removing significantly either gross alpha activity in which uranium was the main contributor (Camacho et al., 2010) or gross beta activity (produced by K-40 activity).

After introduction of RO the mean reduction of gross alpha and beta activities were 38 % and 37 % respectively, for the period 2011 to 2013. These values are below those published by our group (Montaña et al., 2013) for this facility since as it has already been mentioned, the treatment plant just treat a certain percentage of the water depending operational conditions.

As regards the long-term radiological study some statistical parameters are presented in Table 1.

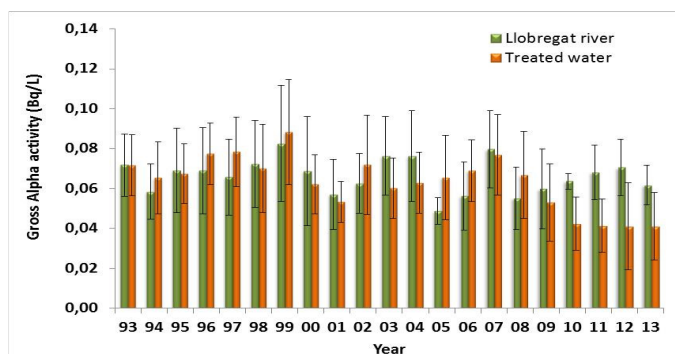


Figure 1. Temporal evolution of gross alpha activity in the Llobregat River and in treated waters in the period 1993-2013.

Table 1. Statistical parameters in period 1993-2013.

Activitiy	Sample (data)	Average $\pm$ standard deviation	Minimum-maximum
Gross alpha	Llobregat river (212)	0.067 $\pm$ 0.019	0.030-0.130
	Cornellà aquifer (76)	0.073 $\pm$ 0.020	0.030-0.145
	Sant Feliu aquifer (52)	0.068 $\pm$ 0.022	0.033-0.130
	Treated water (213)	0.062 $\pm$ 0.021	0.015-0.130
Gross beta	Llobregat river (232)	1.11 $\pm$ 0.33	0.45-2.81
	Cornellà aquifer (94)	1.17 $\pm$ 0.25	0.74-1.84
	Sant Feliu aquifer (62)	1.11 $\pm$ 0.28	0.82-2.12
	Treated water (239)	0.98 $\pm$ 0.32	0.37-1.85

No significant differences were found between average radiological values in samples from the aquifers, from the Llobregat River and from treated water. Maximum values both in gross alpha and beta activities were detected before 2000 when occasional short-term breakages of a brine collector caused salt concentration peaks in the river.

The conventional treatment used did not reduce gross alpha and gross beta activities. However, the membrane techniques used by RO did produce a significant decrease in radioactivity levels.

In the temporal study no relevant trend was identified in samples from the aquifers or from the Llobregat River after year 2000.

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# Time series analysis of long term (1988-2015) gamma radiation measurements in Thessaloniki , Greece.

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Keywords: gamma radiation, portable HPGe, time series analysis  
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The early warning stations, scattered all over Europe, is the best way against any radiological incident that may occur. The usefulness of the data they provide isn't constrained to an early warning, but opens a wide range of data analysis. The amount of information gathered is enormous and more specific for our laboratory, the Nuclear Technology laboratory of Aristotle University of Thessaloniki "AUTH" , the database extends to 26 years with almost 5000 values. The way of making the stations more secure and precise for any future incident is to understand and analyze the past, and that is exactly the purpose of the present work.

The Gamma radiation measurements were performed during the last 26 years, starting from 1988, with a NaI detector (Xetex system) outside the Laboratory and a time series was created. Measurements were also performed in the same place during 1995-1998 and 2014-2015 with portable HPGe detector (Figure 1).

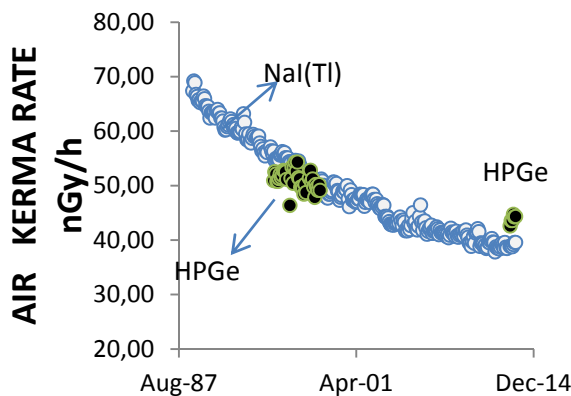


Figure 1. Mean monthly air kerma rates measured with NaI(Tl) and portable HPGe detectors

The total gamma air kerma rate decreases exponentially with time. The total air kerma rate is the sum of the air kerma rates due to 1) Uranium series 2) Thorium series 3) K-40 4) Cs-137 (due to the Chernobyl accident). From the time dependence measurements with the HPGe detector it was found that the time dependence air kerma rates due to 1) Uranium series 2) Thorium series 3) K-40 is quite constant . On the contrary , the exponential decrease of air kerma rate due to Cs-137 is stronger than expected

due to the natural decay of Cs-137. Gamma air kerma rates due to Cs-137, decreases exponentially with an effective Half-life ( $t_{1/2}$ ) of about 14 years. This means that air kerma rates decrease with time not only due to the radioactive decay of Cs-137 (Half-life ( $t_{1/2}$ ) of 30 years) but also due to a "removal" of Cs-137 from the field of view of the detector.

Time series analysis of the mean monthly total gamma air kerma rates was performed. Applying Fast Fourier Transform (FFT) to the time series was found, as expected, an annual cycle. In addition a 4 year periodicity could be also deduced from the FFT analysis. Applying advanced statistical analysis to the time series (like Zhao Atlas Marks Transform) it was possible to find the dynamic (in time) evolution of the periodicities. ZAM maintains both the time information such as the frequency domain , showing changes of the dynamic behavior of the phenomenon being examined through measurements. It was found that the 4 year periodicity is very strong during the years 2002-2004 and 1996-1998 (Figure 2).

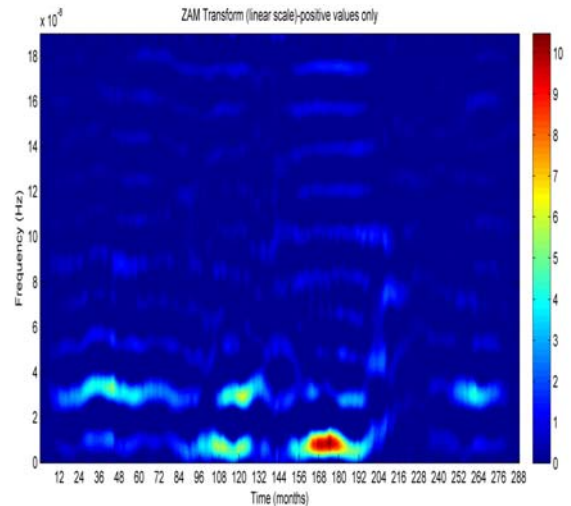


Figure 2.: Zhao Atlas Marks transform of the mean monthly total gamma air kerma rates. The first month corresponds to September 1988. The 4 year periodicity is very strong (in red) during the years 2002-2004.

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## PHOENIX: A web-based platform for accessing and evaluating Hanford Site groundwater contaminant data

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PHOENIX (PNNL-Hanford Online Environmental Information eXchange) is a web-based GIS platform that provides near-real-time public access to current and historical Hanford Site environmental data and integrates it with other relevant Hanford Site geo-spatial data. The Hanford Site, encompassing ~1,500 km<sup>2</sup> in southeastern Washington State, has more than 180 km<sup>2</sup> of groundwater contaminant plumes exceeding drinking water standards. Every year, ~1,300 groundwater wells are sampled, resulting in more than 280,000 distinct analytical results. These operations generate a quantity of data that is challenging for analysts, regulators, and stakeholders to access, visualize, and interpret in a timely manner. Furthermore, these data reside in multiple databases separated by technical or contractual barriers, and have not been made easily accessible to the public, stakeholders, or regulators.

Pacific Northwest National Laboratory deployed PHOENIX to provide intuitive and immediate access to the most current information on the Hanford environment and cleanup conditions, particularly site-

wide groundwater monitoring results. PHOENIX integrates and displays these data geo-spatially on a map with other geo-spatial data, providing important context to the results and revealing previously undiscovered correlations. Additionally, the PHOENIX platform can host analytical and geoprocessing tools and models.

PHOENIX realizes broad, public accessibility by running in common web browsers without the need for specialized software or database access protocols. It accommodates a wide range of users with an intuitive interface that enables users to quickly obtain and visualize data with little or no training. This ease-of-use prompted the U.S. Department of Energy to replace the cumbersome paper-based Hanford Annual Groundwater Monitoring Report in 2012 with a PHOENIX-based online version that provides the same content but also enables users to explore all of the data underlying the report. PHOENIX decision support capabilities have been further enhanced by integrating other Hanford data sets such as vadose zone monitoring, in-tank monitoring, and soil measurements.

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# Testing Voxel Models: Voxelized vs. Ellipsoidal Rabbit Models for use in Radiological Dose Rate Calculations

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Keywords: voxel models, ERICA, MCNP  
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Two voxel models were created for this study, one of an adult rabbit (Figure 1), and one of a juvenile rabbit. These models were used to test assumptions regarding the utilization of such models for radioprotection of the environment. Specifically, voxel models have raised questions regarding variable tissue composition and density effects on absorbed fraction (AF) values, along with implications for age-dependent dose rates as organisms mature.

AFs for electron energies of 0.1, 0.2, 0.4, 0.5, 0.7, 1.0, 1.5, 2.0, and 4.0 MeV and photon energies of 0.01, 0.015, 0.02, 0.03, 0.05, 0.1, 0.2, 0.5, 1.0, 1.5, 2.0, and 4.0 MeV are provided for eleven rabbit tissues. Consistent with previous models, this work demonstrates that AF results are primarily influenced by target organ mass and geometry relative to the source.

Also presented are dose conversion factors (DCFs) for both the adult and juvenile rabbit for C-14, Sr-90, Po-210, Co-60, Cl-36, Ra-226, Pu-239, Cs-137, U-238, Th-232, Pu-240, Am-241, I-131, and U-235. This data was then used to calculate organ dose rates for rabbits and other small burrowing mammals at Maralinga in Australia, the Nevada Test Site in the USA, the Hanford site in the USA, and a contaminated forest outside the Fukushima nuclear power plant.

Dose results for rabbits elucidated in this study indicate that varying tissue composition between two mammalian tissue types (e.g. human vs rabbit bones) made little difference in SAF values (within 5% over most energy ranges). However, variable tissue density (e.g. bone vs liver) can significantly impact SAF values. An examination of differences across life-stages revealed increasing self-AFs (SAF) with testis and ovary size of over an order of magnitude for photons and several factors for electrons, indicating the potential for increasing dose rates to these sensitive organs as animals mature.

A ratio of the voxelized rabbit DCFs to those from an ellipsoidal shape of the same approximate size and mass (created in ERICA) demonstrated that ellipsoidal models are conservative in the vast majority of cases, and are generally fit for regulatory purposes (Ruedig et al. 2014; Gomez-Ros et al. 2008; Brown et al. 2008). However, voxel models may be useful when performing detailed dose-effects studies, as they account for organ crossfire doses, e.g. skeleton irradiating gonads. Additionally, voxel models can more accurately determine dose rates to sensitive tissues.



Figure 1: Rabbit Model

## Acknowledgments

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Eyrolle-Boyer F. et al (2015) Tritium and  $^{14}\text{C}$  background levels in pristine aquatic systems and their potential sources of variability, *Journal of Environmental Radioactivity*, 139, 24-32.

# Selection of phyto/phyco-remediation systems for radionuclide decontamination

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Keywords: algae, cyanobacteria, environmental conditions, higher plants, radioactive contamination.

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Although there are various studies dealing with the application of plants, algae, and cyanobacteria for the decontamination of certain radionuclides from aqueous solutions, no review exists that compares these different approaches to identify the most promising decontamination scheme for scenarios related to nuclear installations. In addition, these studies are often conducted under specific environmental conditions without knowing to what extent the extraction capacity might depend on the experimental conditions.

The goal of this research is to assess the potential of using cyanobacteria, micro/macroalgae and higher plants for the remediation of radioactively contaminated water within different scenarios related to nuclear installations.

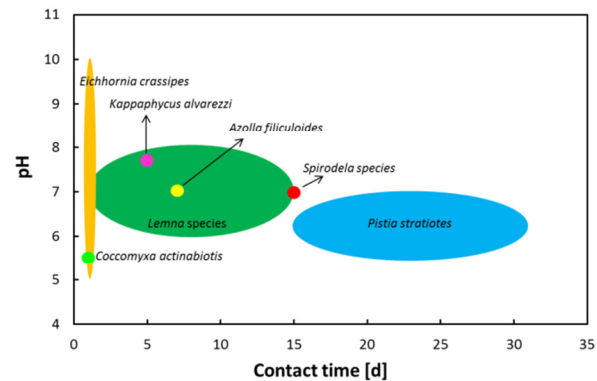
A review is made of the existing literature on plant and algae based water decontamination systems for radionuclides relevant for contamination scenarios related to nuclear installations (e.g. <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am, <sup>58</sup>Co, <sup>60</sup>Co, <sup>110m</sup>Ag, <sup>54</sup>Mn, <sup>59</sup>Fe, <sup>51</sup>Cr, <sup>56</sup>Zn, etc.). In a first phase, the selection of potential organisms was made based on selection criteria such as plant/biomass concentration, maximal removal and contact time. Subsequently, the environmental conditions which allowed phyto/phyco-remediation were described and the most influencing or restricting environmental parameters identified. A difference was made between bioaccumulation systems in which elements are actively taken up and transported into the cell and biosorption systems in which elements are attached to the cell wall by physical adsorption or ion exchange.

For the bioaccumulation systems we considered living micro/macroalgae, terrestrial plants and aquatic plants. The flexibility of these systems is represented in figure 1 for a limited number of elements/radionuclides and organisms. The most influencing/restricting environmental parameters were pH, amount of biomass, contact time, and competing ions. It was shown that the optimal pH was highly dependent on the plant species and element considered (Saleh, 2012). In addition, Sr<sup>2+</sup> uptake is reduced in the presence of Ca<sup>2+</sup> while Cs<sup>+</sup> uptake is restricted in the presence of K<sup>+</sup> (Soudek *et al.*, 2006).

For biosorption systems (living cyanobacteria and microalgae and the dead biomass of cyanobacteria, algae and plants), again figures were made that show the flexibility of the biosorbents. The most influencing environmental parameter was pH (especially for dead biomass) (Pohl, P. and Schimmack, 2006). In addition, several studies reported on the possibility to improve the performance of the biosorbents by chemical modification of dead biomass (Popa *et al.*, 2006). Biosorption systems have the major advantage (compared to plant

systems) that the necessary contact time is lower (hours compared to days). When using dead biomass, some additional advantages can be listed such as the possibility to optimize performance by chemical treatment, no culture maintenance requirements, the possibility for desorption to reuse biomass, etc.

A.



B.

	Co	Cr	Ag	Mn	Sb	Fe	Zn
<i>Azolla filiculoides</i>		?					?
<i>Cocomyxa actinabiotis</i>	91		100		30		100
<i>Eichhornia crassipes</i>	90	85		?		90	95
<i>Kappaphycus alvarezii</i>	?						
<i>Lemna species</i>	85	97		95		98	98
<i>Pistia stratiotes</i>		99		98		95	92
<i>Spiradela species</i>		83		97		84	96

Figure 1. (A) Contact time and pH ranges under which the indicated living plants/algae were tested and shown to work for the elements given in (B); (B) Overview of the maximal removal percentages obtained from literature

This study shows the high potential for various organisms/biomass-types as remediators of (mixtures of) radionuclides in contamination scenarios related to nuclear installations. From the evaluated organisms, aquatic plants, microalgae, and cyanobacteria are identified as the most promising organisms to remove radionuclides from aqueous solutions.

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# European-scale investigations on radionuclides in the atmosphere: Knowledge improvements consecutive to the Fukushima accident

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Keywords: aerosol, resuspension, fog deposition, size distribution, gaseous iodine

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The Fukushima accident features (scenario, location and time of the year) revealed several topics of interest regarding the initial behavior of radionuclide-labeled aerosols as well as long post-accident and secondary emissions from contaminated areas.

Regarding the initial deposition conditions, it should be noted that the accident took place end of winter. It is likely that most discrepancies between field measurement and model assessments can be ascribed to snow deposition and fog deposition, especially after the release from unit 2 on March 15. Past studies had already identified rainout scavenging as a major contributor for wet deposition. However there is still a lack of information about the fog capability to make significant deposition of radionuclide-labeled aerosols acting as cloud condensation nuclei or when interstitial aerosols are scavenged by falling or suspended droplets. Snow scavenging remains quite misunderstood but at least as efficient as rain while there exists probably a large range of snowout coefficient values, depending on the snowflake shape, number per unit volume, relative humidity and aerosol number and size.

Again for the short term consequences, it is of primary importance to have a better insight into the size distribution of aerosols in case of an accident and especially at short and medium distances, for dry deposition modelling assessment or for refined assessment of inhalation dose estimates. Some <sup>137</sup>Cs-rich particles were found 180 km from the damaged plant with diameter up to 2-3 µm. Apart from accident conditions, size distribution studies require specific high volume samplers that are rarely designed for sizes below 0.3-0.4 µm. It has been demonstrated that secondary emissions from contaminated areas in Japan was characterized by a large contribution from these small fractions even if the coarse fraction was also of concern. This observations refers both to wind resuspension (usually responsible for coarse particles but that can produce smaller particles after sand-blasting) and also to biogenic aerosols. Pollens correspond to coarse aerosols but there exist other biogenic aerosols that are expected to play an important role in the long term persistence of airborne radionuclide, such as wax exfoliation or biomass burnings and possibly yeast and bacteria emission.

Regarding the released radionuclides of major concern attention should be paid to iodine and variation of the gaseous/particulate ratio over time. There are still some questions to be solved about the transfer rate of gaseous iodine onto particles (adsorption) and possible desorption after deposition. We show that the AMAD (aerodynamic median activity diameter) of aerosol carrying <sup>131</sup>I did not decrease as much as for Cs isotopes. This result from the gradual transfer of gaseous <sup>131</sup>I onto particles of all sizes met during transport and whose coarse fraction is rapidly deposited.

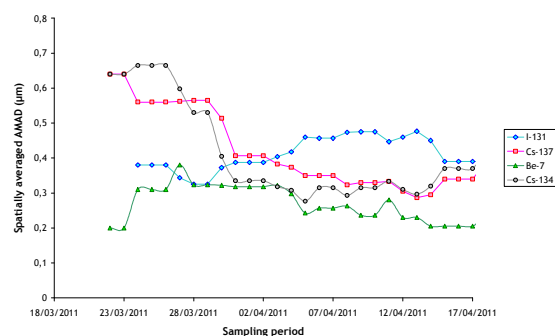


Figure 1: European spatially averaged AMAD time trend for <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>131</sup>I

Response to iodine determination at European scale revealed that, even if it remained of no concern for public health in Europe, the capability to attest for the gaseous iodine presence was rather limited compare to the number of aerosol sampling equipment and determinations. Most attention was dedicated to Cs isotopes and particulate iodine while the main activity levels came from the gaseous <sup>131</sup>I

Some European collaborations linked to Fukushima-labeled radionuclides were performed by the Ring of Five (Ro5) network. This informal network dedicated to trace levels in the atmosphere on the European scale made it possible to investigate arrival date, magnitude and time series of airborne radionuclides; their size distribution and distribution of activity levels along the vertical. Based on these fruitful collaborations, it was decided to structure this network in a proactive manner by highlighting other questions or topics of interest and share knowledge, equipments and methods to deal with these issues.

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# PRODUCTION OF IAEA CERTIFIED REFERENCE MATERIALS FOR RADIONUCLIDES IN MARINE ENVIRONMENT STUDIES

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**Keywords:** Reference Materials, Certified Reference Materials, Radionuclides, Marine Environmental, Accreditation

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Accurate and precise determination of radionuclide activity concentrations in marine samples is important for marine radioactivity assessments and for the use of radionuclides in the study of oceanographic processes. To address the problem of data quality, the IAEA's Environment Laboratories (IAEA-EL) in Monaco have conducted inter-laboratory comparison exercises on radionuclides in marine samples for almost fifty years as part of their contribution to the IAEA's programme of Analytical Quality Control Service (AQCS), currently named as "IAEA's Reference Products for Science and Trade" (Povinec and Pham, 2001, Sanchez-Cabeza et al, 2008). An important part of this activity was the production of Reference Materials (RMs), which were in previous time's products of worldwide inter-laboratory comparison exercises.

The IAEA's Reference Products for Science and Trade programme for radionuclides in the marine environment has recently focused on the production of Certified Reference Materials (CRMs) (ISO 35; Pham et al, 2014; Povinec et al., 2007) with the aim to improve the accuracy and precision of analyses carried out by the laboratories and thus the quality of data, and to provide traceability to SI standards. At present the characterisation of CRMs is being upgraded with accreditation following ISO 35, ISO 34 and ISO/IEC 17025.

An overview of the past, present and future production of CRMs for radionuclides in support of marine environmental studies is presented and discussed. CRMs are valuable standards for method development and validation: they can indicate the need to improve or change existing methods and/or the need of further training. In fact, reference methods should only be accepted on the basis of inter-laboratory comparison tests performed on selected CRMs. RMs and CRMs for all important marine matrices, such as sediment, biota,

sea water, seaweed... etc were produced by the IAEA and are available to the scientific community in research and monitoring laboratories in Member States.

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# Radiological Environmental Aspects of High-Power Accelerator Facilities: Can They Be Showstoppers?

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*Keywords: radiation protection, environment, public, accelerator.*

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With the development of the accelerator technology, especially in the last decade, very powerful beams of strongly interacting particles like protons or heavy ions are available. The sole objectives of increasing the intensities are attaining a high statistical significance of experimental results or observing very rare processes, like interactions of neutrinos or discovering other weakly interacting particles, like the dark-matter candidates.

At present, for fixed-target experiments, the beam power can range from a significant fraction of a MW till several MW exploited during a substantial fraction of a year (e.g. nine months).

For colliders, the energy stored in one circulating beam reaches hundreds of MJ (for example the *nominal* energy per beam in [LHC](#) is 530 MJ equivalent to a 53-ton track falling from a height of 100 m). Such energy is dumped or partly consumed in non-superconducting elements of the facility more than once a day during long periods of an operational year.

Examples for the high-power fixed-target experiments are facilities operated in the past, like [CNGS](#) (CERN Neutrinos for Gran Sasso), or future projects like the [ESS](#) (European Spallation Source), [CENF](#) (CERN Neutrino Facility), [LAGUNA-LBNO](#) (Long-Baseline Neutrino Oscillations) and [SHIP](#) (Search for Hidden Particles).

The powerful hadron colliders include the already shut down [Tevatron](#), [RHIC](#) (Relativistic Heavy Ion Collider), LHC (Large Hadron Collider) in its Run 2 and the [FCC](#) (Future Circular Collider) project that will shape the R&D in the high-energy physics accelerator technology in the following decades with all spin-outs it may bring.

The radiological environmental aspects of hadron accelerators were succinctly described in (Vojtyla, 2009). Basically, there are four aspects: (1) Stray radiation in the environment; (2) Releases of activated air into the atmosphere; (3); Discharges of activated or contaminated water; (4) Soil and groundwater activation.

It has soon turned out that a tendency to re-use existing infrastructure for new or substantially upgraded facilities to save material and human resources has its limits, namely because accelerator sites used to be situated in densely populated areas with a lot of offices, dwellings and agricultural zones located on the sites or immediately around. A challenge has arisen for environmental modellers to cope with situations that are not standard. For example, instead of using static and simple Eulerian atmospheric dispersion models, well working for nuclear power plants, computational fluid-

dynamics models in the environment are tested at CERN at present; starting with accident scenarios for risk analyses.

The best example is the upgrade of the Proton Synchrotron at CERN during the Long Shutdown in 2013–2014. Since its conception 50 years ago, the machine increased its beam intensity by three orders of magnitude and had not fulfilled the justification and facility-optimization obligations of the present radiation protection regulations anymore (Bruno et al., 2011). Additional shielding and a new ventilation system had to be installed.

The project managers as well as the chief executive officers of accelerator laboratories became aware of the situation and radiological environmental impacts, as well as other Safety issues, are considered already during the conceptual design studies of new facilities or before essential upgrades of the existing ones.

The paper gives an overview of the solutions mitigating the four aspects listed above, namely (1): Construction of the facilities deep underground with access shafts formed in chicanes to minimize the leak of neutrons out (sky-shine); (2) Staged and partially-closed ventilation systems of hot areas; (3) Retention basins and HTO-evaporation plants for water discharges; (4) Geotechnical and hydrogeological studies carried out before the civil engineering design starts.

Sometimes, it may happen that a site foreseen for a facility is unsuitable due to environmental constraints and a project must be abandoned, as no solution can be found for reasonable costs.

The magnitude of the problems is illustrated on the example of the FCC – the absolute challenge in the accelerator technology at present. The machine shall circulate two 50 TeV proton beams (8400 MJ/beam) in a tunnel of 80–100 km in circumference buried as deep as 800 m underground and passing limestone areas known as good aquifers (drinking water reservoirs) at some places. It can be compared with the LHC in its *Run 2*: two 6.5 TeV proton beams 75–150 m underground.

Unexpected challenges may appear, e.g. activation of heat-transfer fluids in geo-thermal energy facilities or long-range secondary muon radiation potentially emerging at the ground level.

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## Radiological surveillance of aerosol and fallout in France: last improvements in terms of sampling and measurement

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The OPERA-Air (Observatoire PERmanent de la RADIOactivité de l'Air) sampling network is part of the environment radiological surveillance handled by the French Institute of Radioprotection and Nuclear Safety (IRSN) in France. This network settled in 1959 is aimed at the characterization of airborne trace level. This lasting achievement was possible thanks to many technical and methodological improvements that allow monitoring of decreasing levels of man-made radionuclides. Today it includes, in addition to 40 low flow (from 10 to 80 m<sup>3</sup>.h<sup>-1</sup>) samplers, 8 high and very high flow (namely 300 and 700 m<sup>3</sup>.h<sup>-1</sup>) aerosol samplers. This network also comprises rain and cloud water samplers in order to observe and study the atmospheric wet deposition processes.

This paper will focus on the samples measured by low level gamma ray spectrometry in the laboratory of environmental radioactivity measurement (LMRE): rain, cloud and fog water samples and aerosol filters taken by the high flow and very high flow samplers.

The last developments made both regarding sampling and preparation of the atmospheric samples will be described. The improvements on the air samplers with higher flow, larger surface or thinner filters have been carried out to obtain a larger amount of radioactivity to measure as well as a smaller counting geometry for better detection efficiency.

All these efforts on sampling and preparation went in concert with the metrology improvements, with new measurement equipments such as low background detectors, well-type detectors or anti cosmic set ups. The developments made during the last decades to improve the detection limits, and particularly the efforts to decrease the background and increase the detection efficiency will be described.

All these implementations were and are still made in order to face the decreasing anthropogenic radioactivity levels in the environment, especially in its atmospheric part, and to keep the OPERA-Air

network meeting its dual purpose of surveillance as well as research.

Some results obtained thanks to the sampling, preparation and measuring improvements will be presented for illustrative purposes:

- Determination of <sup>137</sup>Cs at trace level in fog/clouds waters in order to study the rainout scavenging mechanism and to improve the capability in radionuclide deposition modelling.
- Determination over France of trace elements at a few µBq.m<sup>-3</sup> during the Fukushima Dai-ichi accident such as <sup>129m</sup>Te-<sup>129</sup>Te, <sup>132</sup>Te-<sup>132</sup>I, <sup>136</sup>Cs and <sup>140</sup>La.
- Detection of <sup>134</sup>Cs due to the Fukushima accident in rain water samples until November 2012 giving information on the scavenging efficiency of precipitations below the cloud base (washout mechanism) in addition to the rainout mechanism.
- Detection of <sup>131</sup>I and <sup>125</sup>I traces in the air due to abnormal releases from a Hungarian facility.
- Determination of the size distribution of airborne radionuclides soon after the Fukushima Daiichi nuclear accident in France or during normal operation in the vicinity of French nuclear facility in order to improve the modelling capability and deposition calculations.



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# Determination of the Global Coverage of the CTBTO IMS Xenon-133 Component for the Detection of Nuclear Explosions

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**Keywords:** *radioxenon, CTBTO, nuclear explosions.*

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Radioxenon is an important atmospheric tracer to detect underground nuclear explosions. The Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) International Monitoring System (IMS) is designed to provide worldwide continuous physical monitoring and detection of nuclear explosions and incorporates 40 noble gas monitoring stations. They are constantly sampling the atmosphere for concentrations of radioxenon. This work analyses how effectively the network of stations is able to detect unusual xenon-133 concentrations in the atmosphere. A large multitude of nuclear explosions, evenly distributed in space and time, is simulated and the detection rate is calculated. Atmospheric transport modelling is applied to calculate the source-receptor-sensitivities for each monitoring station. The approach includes the anthropogenic radioxenon background, station-specific detection criteria, different scenarios for surface and subsurface nuclear explosions, and a spatial as well as a time dependent analysis. Recommendations are drawn for the improvement of the detection capability:

- the IMS noble gas stations have been distributed more or less evenly across the globe and as a result do not account for the meteorological transport processes in the equatorial region. Therefore, blind spots exist, lowering the detection probability of nuclear explosions;
- furthermore, the distribution of legitimate radioxenon emitters have clearly a negative, but more regional than global, impact. This means that an Isotope Production Facility (IPF) can noticeably decrease the coverage in downwind regions, but the net impact on the global coverage of the Earth's surface including the oceans is minor. However, due to the economic nature of the medical isotope market the number of emitters and their releases will certainly change over time. Accordingly the impact on the detection capability will change regionally;
- it is of significant interest for an effective and stable noble gas detection capability to reduce or at least maintain the existing background, which is mainly created by IPFs. In the interest of the Treaty it is desirable that new facilities follow a proposed emission

maximum, e.g., 5 GBq/day. This guideline is easier to implement for upcoming facilities than for existing ones. The latter have little reason to make costly changes to already state-approved processes within their facility, which do not stand in contradiction to any respective law;

- further improvement is possible by better understanding the noble gas background and more sophisticated detection criteria, e.g., including isotopic ratios and/or time series analysis;

- at the moment the biggest enhancement of the noble gas component could be gained by adding more stations in certain equatorial regions.

## Acknowledgments

The authors gratefully acknowledge the support by the National Scientific Committee Technology of National Institute of Nuclear Physics for the Environmental Radioactivity Monitoring for Earth Sciences (ERMES) project. The authors are grateful to the computer lab of the Italian National Institute of Nuclear Physics and to the Department of Mathematics and Physics of the Roma Tre University. Station and sampling data have been kindly provided through the virtual Data Exploitation Centre (vDEC) within the framework of an agreement between the CTBTO and the Department of Mathematics and Physics at Roma Tre University. The atmospheric transport modeling has been accomplished with meteorological data provided by the European Centre for Medium-Range Weather Forecasts under a collaboration agreement with the Department of Mathematics and Physics at Roma Tre University.

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# Are cosmogenic Be-7 and Na-22 or artificial isotopes (legacy of atmospheric nuclear weapon tests) tracers of stratospheric inputs in groundlevel air?

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We report measurements of weekly Be-7, Na-22 and Cs-137 concentrations in aerosol samples collected with, high volume aerosol filters (HV) at 5 sampling sites in Switzerland from 1994 to 2013 (Be-7, Cs-137) and from 2000 to 2013 (Na-22). For the time period from December 2011 to October 2013 we modelled the origin of air masses for all sampling sites in order to compare whether certain isotope patterns might be correlated with intrusions of stratospheric air. The model used was ECHAM-HAM. The observed variations of the Be-7 and Na-22 activities together with the changes in the Be-7/Na-22 ratio suggest a higher mixing ratio of stratospheric air between March and May. This interpretation, however, is not in line with modelling results of stratosphere-troposphere exchange (STE) showing that the most intense STE occurs during winter. Another possible tracer for stratospheric inputs might be Cs-137, which still exists in the stratosphere due to the atmospheric tests of nuclear weapons. Again, the

modelling data are not in line with this interpretation of the observed Cs-137 peaks during winter.

On a longer time scale, the 11-year solar cycle steering the production of cosmogenic Be-7 is clearly reflected in the Be-7 concentrations in ground-level air. This is illustrated with the annual averages of Be-7 of the Swiss HV sites which correlate very nicely with the cosmic rays measured at Jungfraujoch. Cosmogenic radionuclides in ground-level air thus prove to be valuable tracers of cosmic ray intensity and solar activity on annual scale and are influenced by atmospheric processes only on time scales of seasonal and shorter.

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## Be-7 in various environmental media in the Bremen Region

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### Introduction

The cosmogenic isotope  $^7\text{Be}$  is produced by spallation reactions of cosmic rays with nitrogen, oxygen and carbon in the stratosphere and upper troposphere. It is rapidly adsorbed onto aerosol particles and transported to Earth's surface by wet and/or dry deposition. Our work is focused on quantifying the  $^7\text{Be}$  concentration in different media (air, rainwater, sediments, and soil) for the period of Jan. 2013 until Sep. 2015 with the goal of better understanding the processes of atmospheric mixing, sedimentation and erosion. Additionally several other related isotopes will be used, which are regularly found in rainwater samples, such as  $^{210}\text{Pb}$  and  $^{212}\text{Pb}$  (from individual rainfall events), and in sediment ( $^{40}\text{K}$  and  $^{137}\text{Cs}$ ).

### Experimental

Rainfall is collected since Jan. 2013 on the roof of a campus building, where also a weather station is installed. Sediment is collected since Aug. 2014 in a small stream crossing the campus and air data are provided by the aerosol filter DWD station in Bremen for the total sampling period. All samples are collected on weekly basis. Additionally, for the period Jan. 2013 to Dec. 2014, 10 soil samples were collected routinely in the Bremen area. All samples were measured in wet state in high resolution, low background gamma spectroscopy by high purity germanium detectors of 50% relative efficiency. Soil and sediment are dried afterwards, whilst the rainwater samples undergo volume reduction for the  $^{210}\text{Pb}$  determination. Spectra are evaluated using the Genie-2000 software by Canberra Industries.

### Results and discussion

#### $^7\text{Be}$ in rainwater and air

The seasonal variations in the concentration of  $^7\text{Be}$  in surface air is a well known and studied phenomenon. In the region of Bremen during the study time it ranged between 1-5 mBq/m<sup>3</sup>. Even more pronounced was the seasonal variation of the  $^7\text{Be}$  depositional flux, illustrated in figure 1, with highest values in spring/summer and lowest in winter (0.8- 124 Bq/m<sup>2</sup>).

A clear correlation was found between weekly rainfall amount and  $^7\text{Be}$  surface deposition ( $r=0.73$ ).

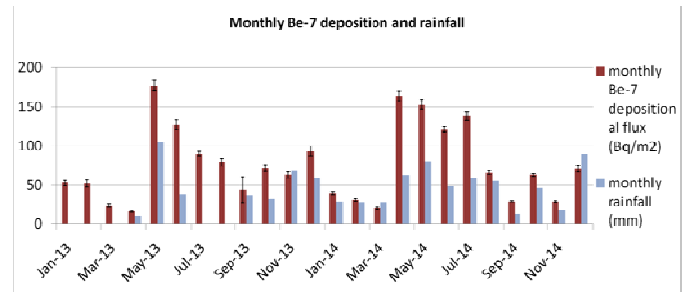


Fig. 1: Seasonal variations in  $^7\text{Be}$  deposition and rainfall for the period Jan. 2013 to Dec. 2014. Missing rainfall data for Jan., Feb., Mar., Jul. and Aug. 2013.

The mean of monthly averaged values of washout ratio, defined as:

$w = \rho_{\text{air}} \frac{C_{\text{rain}}(\frac{\text{Bq}}{\text{kg}})}{C_{\text{air}}(\frac{\text{mBq}}{\text{m}^3})}$ , with  $\rho_{\text{air}} = 1.2 \text{ kg/m}^3$ , the mean air density at 20°C and 0.76m Hg pressure, is calculated as 766 and the deposition velocity is equal to 1.03 cm/s, calculated as:

$$v_d = \frac{F(\text{mean deposition rate } [\frac{\text{Bq}}{\text{m}^2}])}{C_{\text{air}}(\text{mean air concentration } [\frac{\text{Bq}}{\text{m}^3}])}$$

Both values agree with literature data.

Ongoing measurement and study of  $^{210}\text{Pb}$  and  $^{212}\text{Pb}$  isotopes is expected to give insight in local range atmospheric processes.

#### $^7\text{Be}$ in soil and sediment

The activity concentration of  $^7\text{Be}$  in top soil during the period of 2 years showed slight variations between 3-10 Bq/kg(d.m.), whilst  $^{137}\text{Cs}$  had a broader range of 4-25 Bq/kg(d.m.).

In sediments the  $^7\text{Be}$  weekly deposition was much higher (range: 10-460 Bq/m<sup>2</sup>). In fact a mean difference of 130 Bq/m<sup>2</sup> between the deposited weekly  $^7\text{Be}$  due to rainfall and the weekly  $^7\text{Be}$  deposited in sediments indicates an surface runoff process, especially its high correlation to the total weekly rainfall amount ( $r=0.90$ ). This is also supported by the mean weekly deposition of 2.3 Bq/m<sup>2</sup> of  $^{137}\text{Cs}$  measured in sediments.

#### Acknowledgements

Special thanks to Deutscher Wetterdienst (DWD) for providing aerosol  $^7\text{Be}$  data.

# Sources of $^{137}\text{Cs}$ , $^{241}\text{Am}$ and Pu isotopes in the atmosphere

Lujanienė G.<sup>1</sup>, Byčėnienė S.<sup>1</sup>, Šilobritienė B.<sup>2</sup>, Valiulis D.<sup>1</sup>, Šakalys J.<sup>1</sup>, Povinec P.P.<sup>3</sup>

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**Keywords:**  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , Pu isotopes, aerosol.  
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Activities of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  in aerosol samples collected at three sampling stations in Lithuania during 1993-2015 were analyzed with special emphasis on better understanding of their sources and transport in the atmosphere. High volume aerosol samplers (up to  $6000\text{ m}^3\text{ h}^{-1}$ ) constructed at the Institute of Physics using marine motors AOM 42-2 OM5 have been used during aerosol sampling.  $^{137}\text{Cs}$  activities were measured with HPGe detectors (GEM40P4-76, relative efficiency of 40%, resolution of 1.85 keV (FWHM) @ 1.33 MeV, and GX4018, the relative efficiency of 42 %, and resolution of 1.8 keV @ 1.33 MeV). The radiochemical analyses of Am and Pu were carried out on monthly samples (total volume  $\sim 2.0 \cdot 10^6\text{ m}^3$ ) of aerosol filter ashes (about 30 g). The  $^{241}\text{Am}$  and Pu isotopes were measured after radiochemical separation using a TOPO/cyclohexane extraction and purification using UTEVA, TRU and TEVA resins (100 – 150  $\mu\text{m}$ ) (Lujanienė, 2013). After alpha-spectrometry measurements, Pu was then de-plated from the stainless steel disks and after additional cleaning using UTEVA resins it was analyzed for  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio by ICPMS (ELEMENT-2, Thermo Fischer Scientific).

Long-term observation of radionuclide variations in aerosol samples indicated transport of dust-storm, forest fire products from contaminated regions. The characteristic activity ratios of radionuclides and specific Pu isotope ratios were used for the source identification. Characteristic plutonium atom ratios indicated that a release of plutonium isotopes into the environment after the Chernobyl accident was insignificant. A typical Chernobyl-derived plutonium ratio was detected in aerosol samples collected during the accident. The exponential decrease in the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio down to the global fallout value was found. The different behavior of highly volatile radionuclides was found after the Fukushima accident which resulted in enrichment of ground level aerosol particles by  $^{131}\text{I}$  with respect to  $^{137}\text{Cs}$ . It was found that the simulated activities of  $^{137}\text{Cs}$  in aerosol samples after the Fukushima accident reasonably agreed with the activities in aerosol samples at Vilnius site.

Three-dimensional 13-day backward trajectories (Fig. 1) of air masses which reached Vilnius (54\_420N, 25\_300E) at 850 and 700 hPa height were calculated using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) dispersion model (Rolph, 2011).

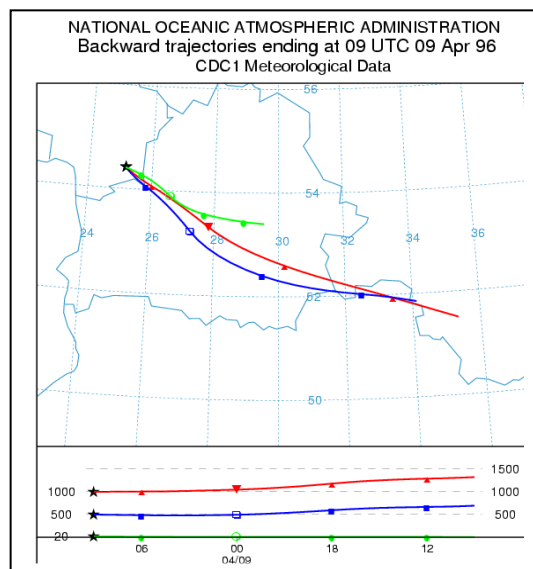


Figure 1. Backward trajectories of air-mass transport of forest fire products transported to Lithuania ending on 9 April, 1996 in Vilnius.

In order to study an association between air mass backward trajectories and the activity concentrations of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  at the sampling site the non-hierarchical clustering algorithm was used. In this study the trajectories were grouped into clusters with similar history, i.e. with a similar path of advection and velocity of air flow. In this way the uncertainties of the individual trajectories were minimized. To calculate the potential areas affecting activities at the site, the potential source contribution function based on the geographic region covering by the backward trajectories was applied.

Parameters for the PSCF model were selected from the Climate Diagnostics Center/NCEP/NCAR Reanalysis archive grid data of the NWS NCEP (for the trajectory duration of 96-h, and the starting height of 100 m).

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# Seasonal variability of $^7\text{Be}$ AMAD and dependency on atmospheric conditions in Athens

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$^7\text{Be}$  is a relative short lived natural occurring radioactive nuclide ( $t_{1/2}=53.3\text{d}$ ) of cosmogenic origin which is formed in the upper troposphere and lower stratosphere by spallation reactions of light atmospheric nuclei of nitrogen and oxygen with cosmic rays.  $^7\text{Be}$  production rate in the troposphere is approximately 33% of the total production in the atmosphere. The  $^7\text{Be}$  produced in the stratosphere, comes into the troposphere by exchange processes due to variations in atmospheric conditions associated with seasonal changes. It then attaches to aerosol particles, which finally deposit on the ground.

Since March, 2011 an atmospheric sampling program by means of High Volume samplers has been established at Demokritos urban background station (GAW-DEM, 2007) located at the North East corner of the Greater Athens Metropolitan Area and at an altitude of 270 m.a.s.l. Measurements were conducted using a six-stage high volume cascade impactor (nominal 50% cutoff sizes: 10.1, 4.2, 2.1, 1.4, 0.73, 0.41, 0.05  $\mu\text{m}$ ). Concentrations of radioisotopes were measured after weekly sampling. Each sample was collected after approximately 84h of sampling (Air Vol. 2500  $\text{m}^3$ ) while since September 2014, continuous weekly sampling was applied (Air Vol 4600  $\text{m}^3$ ). Cellulose Filters and impaction substrates were used (Whatman), cut in a diameter of 70mm. Gamma-spectroscopy was conducted and  $^7\text{Be}$  was detected by its photo peak at 477.59keV. The detector used is a coaxial HP 30% Germanium detector, with energy resolution of 1.93keV for  $^{60}\text{Co}$  (1332keV) and a 8k multichannel analyzer, with energy resolution 0.25 keV/channel. The spectra produced, were then analyzed using the software Interwinner 4.1. In this study,  $^7\text{Be}$  concentrations' time series and parameters from the calculated  $^7\text{Be}$  size distributions were studied with respect to atmospheric conditions during the last 4 years.

Figure 1 displays the  $^7\text{Be}$  seasonal variation with strong temporal variability of  $^7\text{Be}$  atmospheric ground concentrations.  $^7\text{Be}$  maxima are observed during summer months, and minima during winter months, which can be attributed to the stratosphere – to – troposphere exchange process. The amount of rainfall also contributes to the seasonal variation by washout, while significant increase in activity under dry meteorological conditions were due to the non-scavenging of ambient air by rainfall (fig1). The highest activities in the summer months are caused by increased vertical transport of  $^7\text{Be}$  from the upper

troposphere due to decreased stability of the troposphere during the summer months. AMAD ranges from 0.28 to 0.38  $\mu\text{m}$  (avg 0.25  $\mu\text{m}$ ).

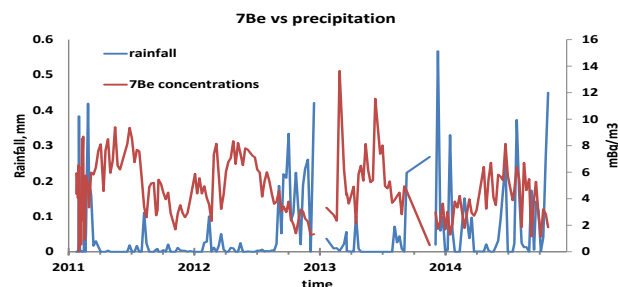


Figure 1.  $^7\text{Be}$  concentrations in air with precipitation

This instability in the atmosphere has an impact on particles' residence time, providing lower AMAD values during summer (Winkler et al., 1998) (fig2).

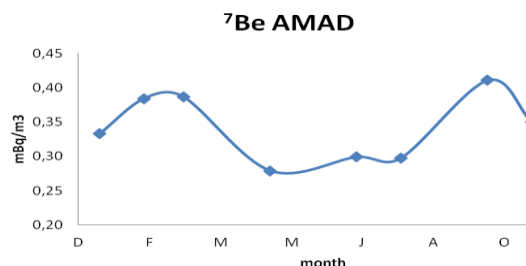


Figure 2.  $^7\text{Be}$  AMAD in the year 2014

This study has proved negative correlation between relative humidity and the activity. This could be explained by an increase in the diameter of airborne particles caused by condensation process, which are more likely to be scavenged, thus reducing activity in the atmosphere (Percot et al., 2013).

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## Gamma emitters in atmospheric precipitation since 2005 in Krakow (Southern Poland)

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*Radioactivity of air, gamma emitters, cosmogenic radionuclides, resuspension.*  
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Radionuclides in atmospheric precipitation are of different origin: soil resuspension, sea spray, industrial emissions, cosmogenic etc. Inputs of all those sources are not constant in time thus resulting content of radionuclides in air precipitation can vary a lot.

Atmospheric precipitation (sum of wet and dry deposition) were collected as monthly samples in Krakow (Southern Poland) since 2005. Samples were collected at stainless steel tray having surface of 2.2 m<sup>2</sup> connected to plastic barrels of 160 L volume (one or two). Water collected within barrels were acidified by adding to it 50 ml of concentrated nitric acid. After collections water was transferred in portions into stainless steel vessel (10 L volume) and evaporated finally to about 1 L. This solution was carefully transferred into a glass vessel. After emptying barrel(s) and stainless steel vessel the walls and bottom of them were carefully wiped using ash-free filtration papers and 1 M nitric acid. Those filters were then dried, ashed in 400 °C overnight and ashes were added to original solution. This solution was again evaporated down to about 100 ml and then it was transferred to plastic vessel (5 cm diameter, 120 ml volume), where it was gently evaporated under infrared lamp. In plastic vessel the collected dust and salts from evaporated water formed dry solid phase layer of typically about 5-8 mm height. Such prepared samples were subject of gamma

spectrometric measurements using 3 different gamma ray-spectrometers, each with HPGe detector and 10 or 15 cm lead shields.

Activity was determined for <sup>7</sup>Be, <sup>210</sup>Pb, <sup>137</sup>Cs, <sup>40</sup>K, <sup>22</sup>Na. Since our treatment can exclude iodine measurements (due to acidic conditions in evaporation) during Fukushima accident time (March-April 2011) samples were treated differently, without additional acidification of water.

The results for 2011 were already published [Mietelski et al., 2014]. For years 2005-2006 radiochemical analyses for Pu isotopes were done and results were published [Kierepko et al. 2009]. The complete ten years record of data for gamma emitters will be presented for the first time during conference.

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## Sources of recent radionuclide variations in the atmosphere

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**Keywords:** anthropogenic radionuclides, atmosphere, resuspension, Sahara dust, biomass burning, dispersion modelling

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Temporal and spatial variations of radionuclides observed in the atmosphere in recent years will be reviewed and the main sources will be identified (Livingston and Povinec, 2002). Radionuclides of cosmogenic (<sup>7</sup>Be), terrestrial (<sup>40</sup>K and <sup>210</sup>Pb) and anthropogenic (<sup>14</sup>C, <sup>137</sup>Cs and <sup>239,240</sup>Pu) origins will be used as proxy for identification of their variations in atmospheric aerosols.

Aerosol and deposition samples collected at the South Europe (Monaco – Pham et al., 2013, and Thessaloniki – Ioannidou et al., 2005), central Europe (Bratislava – Povinec et al. 2012; Sykora et al., 2012, and Krakow – Blazej and Mietelski, 2014), north-east Europe (Vilnius – Lujanienė et al., 2009) and far-east (Japan - Hirose et al., 2008) will be used for identification of the main sources of radionuclide variations.

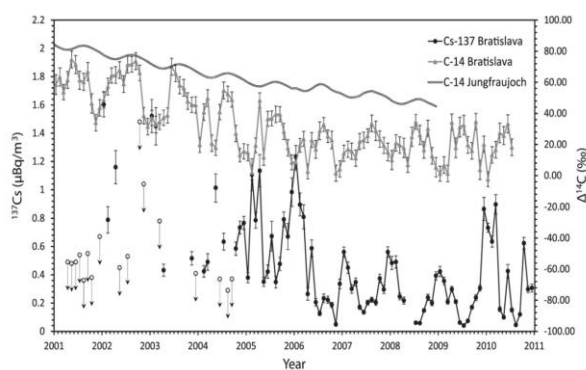


Figure 1. Comparison of <sup>137</sup>Cs and <sup>14</sup>C levels in the Bratislava air.

The main sources of radionuclide variations in the atmosphere before the Fukushima nuclear accident will include: modulation of galactic cosmic rays with solar activity, local re-suspension of radionuclides from soil, desert dust events, biomass burning and volcanic eruptions.

The variation of <sup>137</sup>Cs concentrations in the ground level atmosphere, measured by European monitoring stations, will be correlated with Saharan dust events in which considerably large amounts of dust are transported over the Mediterranean Sea on the European continent. A weather model will be used to simulate a probability of dust uplift from the Saharan desert, and a dispersion model for its propagation through the atmosphere, including dry and wet deposition.

The temporal variations of <sup>137</sup>Cs and <sup>239,240</sup>Pu in deposition samples measured in Tsukuba by MRI (Japan) will be used to study the long-range transport of the Asian dust (produced in the East Asian deserts and arid regions), and its impact on the atmospheric environment in the East Asia (and possibly in Europe).

These studies will help to estimate how large-scale dust events in the deserts and arid regions could affect global atmosphere.

### Acknowledgments

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## Radioactive particles in the floodplain of the Yenisei River

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**Keywords:** radioactive particles; Yenisei River; plutonium complex; dating of particles; sources of high-dose *g*-radiation; bioassays.

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The Yenisei is one of the largest rivers in the world. The Mining-and-Chemical Combine (MCC) of Rosatom is situated on the bank of the Yenisei River at Zheleznogorsk, 60 km downstream of the city of Krasnoyarsk. The Combine, which had been producing weapons-grade plutonium since 1958, consisted of a reactor plant and a radiochemical plant. Two reactors, which used the Yenisei water as coolant for the reactor core, were shut down in 1992. The third reactor was shut down in 2010, but the radiochemical plant is still working. From 1995 to 2014, researchers of the Institute of Biophysics and Institute of Geology and Mineralogy found hot particles during their annual sampling campaigns at the Yenisei (Bolsunovsky and Tcherkezian, 2001; Chuguevskii et al., 2010). These particles were found in floodplain soils and sediments of the Yenisei River in the area affected by the operation of the MCC.

The main objective of this study is to characterise radioactive particles of the Yenisei River and evaluate their potential effects on the River ecosystem using bioassay methods.

Based on comparative analysis of <sup>137</sup>Cs/<sup>134</sup>Cs ratios, all the particles can be divided into three major groups, suggesting that over the 50-year period of the MCC operation, there have been three emergency situations at the MCC reactors, with nuclear fuel microparticles released into the Yenisei.

Radioactive particles of the Yenisei River with the activity of <sup>137</sup>Cs (or another radionuclide) from several kBq/particle to several tens of MBq/particle are a serious hazard to people and biota. Particles that were used in our studies had been found at the riverside in places frequented by local residents. For particles with high activity of <sup>137</sup>Cs (18 to 60 MBq/particle), we calculated exposure doses (mSv/h) from active particles changing with distance. For some of the particles, the exposure dose close to the particles (2 cm) was 3÷11 mSv/h. If a radioactive particle is in contact with skin or other parts of the body, a person may get the annual maximum permissible radiation dose (according to the Russian Federal standards) in a few hours, depending on the distance from the particle.

Living organisms that are actively developing in the Yenisei floodplain soils and sediments and roots of plants and shrubs growing in them are affected by radiation emitted by radioactive particles. In most cases, organisms of the biota receive small radiation doses, whose effects are usually revealed using bioindication

and bioassay techniques. Submerged macrophytes are widely used as bioindicators of contamination of rivers by heavy metals and artificial radionuclides. Cytogenetic studies of elodea (*Elodea canadensis*) growing in the area affected by the MCC effluents showed a higher level of chromosome abnormalities compared to the plants from the reference area (Bolsunovsky et al., 2009). The cytogenetic abnormalities revealed in elodea roots could evidently be caused by the presence of artificial radionuclides, including those of radioactive particles, in sediments.

Environmental toxicity is also studied using bioassays, which are performed in laboratory, with test organisms. In the elodea roots used in the experiment with high concentrations of artificial radionuclides in sediments, the frequency of occurrence of cells with chromosomal aberrations was higher than in the control. In further bioassays, radioactive particles were used instead of radioactive sediments as sources of radioactivity. After 14 days of the experiment with the <sup>137</sup>Cs-particle, the length of elodea roots decreased significantly as the exposure rate increased. In experiments, the percentage of cells of the elodea apical roots that contained various cytogenetic abnormalities in the ana-telophase was dose dependent: as the absorbed dose was increased, the total percentage of cells with chromosomal abnormalities grew, too. Results of the toxicological experiment with radioactive particles as sources of radioactivity showed high sensitivity of elodea to low-dose radiation exposure.

The presence of a large number of radioactive particles in the floodplain of the Yenisei River from the MCC production area and farther downstream is a source of potential hazard to people and biota.

### Acknowledgments

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# Modelling Distribution and Transport of Medical $^{131}\text{I}$ in a Wastewater Treatment Plant

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**Keywords:** wastewater treatment plant,  $^{131}\text{I}$ , transport, modelling.  
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As a result of nuclear accidents,  $^{131}\text{I}$  may be washed out from air or settle onto the ground by dry deposition. In urban landscapes this is again washed out by rain, partly introduced into the sewer system and thus transported to the next wastewater treatment plant (WWTP) where it may accumulate in certain compartments.

On a regular basis,  $^{131}\text{I}$  is released into the urban sewer system in the course of therapeutic and diagnostic treatment of patients with thyroid diseases. This presents an opportunity to use the medical  $^{131}\text{I}$  as a tracer to study distribution and transport processes within a WWTP. In the case of nuclear accidents, an understanding of these processes will be valuable for making prognoses.

In sewage water and sludge from a local WWTP in Bremen,  $^{131}\text{I}$  has been found in three different fractions: (i) as inorganic cation (iodide) in solution, (ii) bound to dissolved organic matter and (iii) attached to suspended particles and colloids. It is shown how these fractions can be estimated experimentally by a sequential extraction method including the precipitation of particulate matter by aluminium hydroxide and subsequent extraction of iodide with Bentonite. The ratios between those fractions measured in samples from various different compartments of the WWTP are compared with modeling results obtained with the geochemical code PHREEQC (Parkhurst and Appelo, 1999).

Figure 1 shows a comparison between the iodine fractions measured at the outflow of the pretreatment unit of a local WWTP in Bremen and a PHREEQC simulation using literature values for waste water composition (Koppe and Stozek, 1999) and constants for iodine binding to particulate (Zhang et al., 2011) and dissolved (Christiansen and Carlsen, 1991) organic matter, respectively. While the percentages for particulate bound iodine are quite similar, the measured percentage ratio of dissolved organic to inorganic iodine is significantly higher than in the simulation. This may be due to underestimation of both the amount of dissolved organic matter (DOM) and the number of binding sites per gram of DOM. This issue will be resolved when DOM measurements from the WWTP pretreatment effluent are available.

More sampling campaigns are under way to measure the iodine fractions at various locations inside the WWTP. This will show whether the  $^{131}\text{I}$  distribution changes on the path of the sewage water through the WWTP and thus be important for transport modelling.

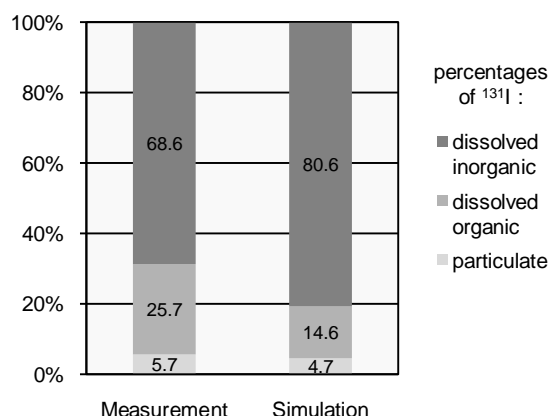


Figure 1. Measured and simulated  $^{131}\text{I}$  distribution in the effluent from the pretreatment unit of a WWTP. The measurement uncertainty is 5% for dissolved organic and anorganic iodine and 2% for iodine bound to particulate matter.

At present, a Simulink<sup>TM</sup> compartment model for the iodine transport through the WWTP is being developed. The model is based on average residence times of water and particles inside a compartment. It will be used for estimating processes of activity accumulation within the WWTP and for predicting the time evolution of the activity of the WWTP effluent after contamination of the inflow.

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# Environmental radioactivity and site-specific concentration ratios of non-human biota in the vicinity of Paks NPP, Hungary

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The planned expansion of Paks Nuclear Power Plant with new units, like every other decision-making on activities, facilities, existing sites and contaminated areas of potential, perceived or actual environmental concern, is governed by an environmental impact assessment (EIA). In accordance with the recent recommendations of the International Commission on Radiological Protection (ICRP) which incorporate direct consideration of the environment (ICRP, 2007), the EIA of this expansion involves the assessment of the levels environmental radioactivity and their effects on non-biota as well. This assessment has been performed with the guidance of the ERICA (Environmental Risks from Ionising Radiation in the Environment: Assessment and Management) Integrated Approach (Larsson, 2008).

The process of estimating exposure of biota, as defined by the ERICA Integrated Approach, involves the estimation or direct measurement of activity concentrations in environmental media and organisms, definition of exposure conditions, and estimation of radiation dose rates to selected biota. Environmental risk is usually quantified to reference organisms (ICRP, 2007). Each reference organism has its own specified geometry (affecting the dose conversion coefficients) and is considered to be representative of terrestrial, freshwater or marine ecosystems. While the activity concentration levels of various environmental medium can be measured directly (in case of existing sites) or estimated using highly sophisticated models (in case of planned facilities) quite precisely, the same is usually not valid to that of the organisms populating the respective ecosystems. In the ERICA Tool (Brown et al., 2008), radionuclide transfer to organisms is calculated using concentration ratios (CRs) in a steady-state fashion, with 'default' CR values readily attributable to the reference organisms. The CR values included in the database were derived from reviews of original publications by preference. However, since existing experimental data can cover a wide range of environmental conditions resulting in usually a wide range of CR values for the same type of organisms, in numerous cases the statistical analysis resulted in relative standard deviations exceeding 100% (or even 1000%!). Consequently, an assessment with Tier 3 of the ERICA Tool (where all sources of uncertainties can be attributed) can easily result in dose rate ranges (e.g. between 5% and 95% probability) covering several orders of magnitude.

With this information in mind, both the base environmental medium (soil, river water and river sediment) and the representative denizens of the ecosystems have been sampled in high definition.

Sample preparation included drying, grinding, combustion, radiochemical separation, etc., and was followed by gamma-, beta- (LSC) and alpha-spectrometry. The new results have been supplemented by the statistical analysis of historical data as well.

An example set of site-specific CRs for strontium and caesium are presented in Table 1, with comparison to ERICA Tool default values (both from versions of Nov 2012 and Nov 2014). Similar sets have been calculated for americium and plutonium as well, and more organisms are covered.

Table 1. Concentration ratios of selected terrestrial and freshwater ecosystem reference organism

Organism	CR	Paks NPP	ERICA Tool	
			Nov 2012	Nov 2014
grasses & herbs	Sr	1.49±1.26	0.21±2.82	0.78±1.62
	Cs	0.023±0.036	0.69±1.08	1.12±2.49
lichen & bryophytes	Sr	1.81±0.50	8.68±7.90	4.64±6.98
	Cs	1.69±1.82	5.61±4.14	3.78±3.81
large mammal	Sr	5.63±2.84	1.74±2.35	1.67±2.29
	Cs	0.004±0.002	2.87±4.25	3.42±8.22
bivalve mollusc	Sr	1250±190	270±95	461±485
	Cs	190±20	460±590	129±103
pelagic fish	Sr	310±55	17±23	860±4800
	Cs	400±50	7100±6000	3370±8060
phyto-plankton	Sr	360±60	40±2	123±122
	Cs	940±690	4700±6500	142±190

As one can see, using the default values instead of the site-specific ones can result in differences of at least a factor of 2 in most cases, especially when compared to the earlier database.

Conclusively, to minimise the level of uncertainties when performing such an assessment, the utilisation of site-specific, directly measured activity concentrations is highly preferred over estimated ones.

## Acknowledgments

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# Exhalation of <sup>131</sup>I after radioiodine therapy Dosimetric considerations based on measurements in exhaled air

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It is well known that a considerable amount of radioiodine is exhaled after radioiodine therapy (RIT) leading to unwanted radiation exposure through inhalation [Schomäcker et al. (2011)].

The exhaled air of 47 patients with different thyroid diseases receiving [<sup>131</sup>I]-NaI were investigated and the chemical iodine species (organic, elemental, aerosol bound) were determined. For different scenarios dosimetric implications were evaluated.

The radioactivity of <sup>131</sup>I in the exhaled air was found between 1 – 100 kBq / m<sup>3</sup> one hour after application decreasing significantly over the time (to less than 1 % of the initially measured activity after 100 h). The fraction of radioiodine in the exhaled air was found between 80 ppm and 150 ppm of the administered activity where organically bound radioiodine represents 94 % – 99.9% [Schomäcker et al. (2011)].

In order to approximate effective doses after incorporation caused by inhalation by persons in contact with the patients different scenarios were considered: Depending on the thyroid disease a monoexponential time-activity curve for the exhalation of radioiodine was fitted to the experimental data and a time-dependent radioactivity concentration  $c_A(t_i)$  in the room was thereby estimated.

$$c_A(t_i) = \frac{RV \cdot \int_{t_i}^{t_{i+1}} c_{ex,0} \cdot e^{-\lambda t} dt}{V_R} \quad (\text{Eq. 1})$$

- RV: respiratory volume per time (1.2 m<sup>3</sup> / h) for exhalation

-  $\int_{t_i}^{t_{i+1}} c_{ex,0} \cdot e^{-\lambda t} dt$  : Exhalation integral (MBq·h / m<sup>3</sup>)

- V<sub>R</sub>: Volume of the room m<sup>3</sup> (assumption: 30m<sup>3</sup>)

Two scenarios were assessed: the first scenario assumed that the patient was discharged from the nuclear medicine ward immediately after administration (“early scenario” = 1). He then stayed with his family eight hours after administration of <sup>131</sup>I for a time period of another eight hours ( $t_i = 8$  h and  $t_{i+1} = 16$  h). The second scenario was that the patient was discharged after 3 d (“late scenario” = 2). Then he “exposed” his family again eight hours later similar to scenario 1 ( $t_i = 80$  h and  $t_{i+1} = 88$  h). Doses were calculated using Eq. 2:

$$E = DF \cdot \sum_{t_i} RV \cdot ARR(t_i) \cdot c_A(t_i) \cdot t \quad (\text{Eq. 2})$$

- DF: dose factor - 8.1•10<sup>-3</sup> μSv / Bq (adults) and 7.2•10<sup>-2</sup> μSv / Bq (infants, 1 year old)

- RV: respiratory volume per time for inhalation (1.2 m<sup>3</sup> / h for adults and 0.252 m<sup>3</sup> / h for infants (1a))

- ARR: air retention rate (assumption: 0.85)

Tab. 1 comprises the mean values of the effective doses calculated for the two scenarios for adults and children:

Table 1. Doses for different inhalation scenarios.

Scenario	Effective doses [μSv] by inhalation		
	Carcinoma	Graves' Disease	Adenoma
Adults (1)	97	31	27
Children (1)	180	57	50
Adults (2)	2	<1	< 1
Children (2)	3	<1	< 1

After immediate discharge the doses received by inhalation may surpass 100 μSv but are significantly reduced in the scenario after three days.

For the “early scenario” maximum doses should also be discussed: In the vicinity of a patient treated for a benign thyroid disease and showing a relatively high exhalation doses can rise up to 125 μSv for adults and 240 μSv for children. After treatment of thyroid carcinoma values can rise up to 270 μSv for adults and 500 μSv for children. On the other hand if the discharge of the patient is three days later in the most cases doses are below 1 μSv. Hence significant doses by inhalation of exhaled radioiodine are to be expected only when the patient is discharged early.

All in all, the effective doses caused by incorporation of exhaled radioiodine will be significantly lower after a period of three days. The inpatient treatment is therefore an important factor for reducing radiation doses to members of the public. The authors of this contribution would not recommend radioiodine therapies as outpatient treatment.

In order to meet the requirement of not exceeding the annual limit of 1 mSv we have to additionally consider the external exposure by patients undergoing radioiodine therapy.

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# Estimation of the geogenic radon potential and radon prone areas from geochemical quantities and ambient dose rate

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Indoor radon (Rn) has been recognized a significant hazard to human health. Therefore it is increasingly being regulated in order to limit exposure to Rn. The latest regulations are part of the new European “basic safety standards for protection against the dangers arising from exposure to ionising radiation” (BSS 2013) which have to be implemented as national law by EU Member States. One task is to delineate Rn prone areas which design regions with elevated probability of encountering high indoor Rn concentrations.

In most cases, the most important source of indoor Rn is the ground below the house. Physically, the relevant quantities are <sup>226</sup>Ra concentration in soil and rock, Rn emanation power and ground permeability which enables Rn transport and eventual infiltration into buildings. Infiltration and accumulation in indoor environments depends on building characteristics and usage types.

The concept of the geogenic radon potential (RP) is to quantify the geogenic compartment of the causal pathway of Rn “from rock to risk”, which can be understood as designating “what earth delivers” in terms of Rn. Quantities which are closest to the concept are combinations of radon concentration in soil air or Ra concentration and the emanation power, and soil permeability. Data of these quantities are available in some European countries but scarce in most.

Therefore, one looks for proxy quantities of the RP which can serve as its predictors, and which are more extensively available. These quantities must be physically and statistically related to the RP.

Two important candidates as continuous quantities are uranium concentration in soil and rock, and ambient dose rate. The former are available through European databases with continent-wide coverage and national datasets. Dose rate is available from the EURDEP system (see references), a network of about 4500 continuously working monitoring stations all over Europe whose purpose is radiological emergency

warning, but which also yield the terrestrial gamma background. A categorical predictor is geology, in terms of geological units defined appropriately as having characteristic RP.

Clearly, these quantities are only imperfectly correlated with the RP. For example, also the thorium series and <sup>40</sup>K contribute to dose rate, but they are not related to the RP. Statistically, imperfect correlation between predictor and predicted quantity leads to uncertainty of the prediction.

In this contribution, we present the datasets and investigate their relations with the RP. We show how, and to which degree of accuracy they can be used for estimating the RP. We also address geostatistical methods for spatial estimation, i.e. mapping.

Geogenic radon prone areas are understood as ones in which the value of the RP meets some criterion, e.g. exceeding a threshold. We discuss how such thresholds could be established by linking the RP with indoor Rn concentration, which is the radiologically more relevant quantity than the RP. Deciding whether a spatial unit is radon prone area or not, amounts to a binomial classification. We demonstrate how this is done by statistical means, and show first maps of the RP and of Rn prone areas on European geographical scale, based on the different predictor quantities. We discuss limitations of our approach and estimation and classification uncertainties.

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EURDEP:

<https://eurdep.jrc.ec.europa.eu/Basic/Pages/Public/Home/Default.aspx>



# Radionuclides distribution in soil grain-size fractions in places of nuclear tests at Semipalatinsk Test Site

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*Keywords: Artificial radionuclide, soil, grain-size fraction, distribution, Semipalatinsk test site*  
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Total 456 different nuclear weapons tests were performed at the Semipalatinsk test site during the period of its operation (1949-1989). Nowadays Semipalatinsk test site is used as unique natural laboratory for scientific studies of artificial radionuclides behavior in all objects of environment.

Distribution of artificial radionuclides in soil grain-size fractions was researched with aim to obtain both scientific knowledge about artificial radionuclides migration in soil components under different conditions of radioactive contamination and actual data for calculation of man inhalation dose.

The research objects were places where atmospheric and surface nuclear tests were carried out as well as plumes from their fallout, sites of underground and excavation tests and also conditionally "background" territories.

Two methods were used for grain-size analysis of soils. For fractions sized particle larger than 40  $\mu\text{m}$  in diameter the wet sieve method was used. For particles sized smaller than 40  $\mu\text{m}$  the sedimentation technique was used.

By results of research, distribution of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  in granulometric fractions of soil has its peculiarities depending on character and mechanism of radioactive contamination. So directly in places of nuclear tests and on plumes definite fractions, significantly enriched with artificial radionuclides are found in soil. In the epicenters of nuclear explosions "enriched" fractions are more often represented by grain-size fractions larger than 500  $\mu\text{m}$ . On plumes of radioactive fallouts depending on distance and yield of explosion enriched fraction get decreased in size.

In addition to presence of "enriched" fraction there is a tendency for natural redistribution of radionuclides in granulometric fractions of soil, expressed in increasing of their accumulation with decrease of fraction size. Natural redistribution can be the most clearly observed for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  if compared with

transuranium radionuclides as  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$ .

At conditionally "background" territories distribution of radionuclides on granulometric fractions of soil mainly depends on their natural redistribution. However in character of distribution of  $^{239+240}\text{Pu}$  we can see the impact from nuclear tests performed at the Semipalatinsk test site.

For example, figure 1 shows  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  distribution on grain-size fractions of soils on plum of surface explosion and adjacent "background" area.

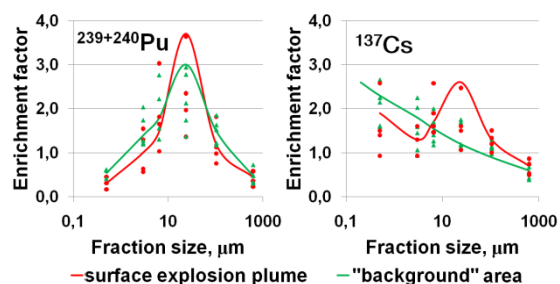


Figure 1. Radionuclides distribution on soil grain-size fractions in different places of radioactive contamination.

So as a result of performed researches the main mechanisms of radionuclides distribution in granulometric fractions of soil in places where nuclear weapons tests were carried out at the Semipalatinsk test site were revealed: "enrichment" of some definite soil fraction, presumably due to presence of radioactive materials of nuclear explosion and natural redistribution of radionuclides due to sorption and other physical and chemical processes in soil.

# Research of species of artificial radionuclides $^{137}\text{Cs}$ , $^{90}\text{Sr}$ , $^{239+240}\text{Pu}$ и $^{241}\text{Am}$ in soils of conditionally «background» territories of Semipalatinsk Test Site

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**Keywords:** Semipalatinsk Test Site, conditionally background territories, artificial radionuclides, speciation of radionuclides in soil, sequential extraction  
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The work provides results of research of species of artificial radionuclides in soils of conditionally «background» territories of the STS. These territories include «northern», «western», «southeastern» and «southern» parts of the STS, specified in this way according to their geographic location at the test site territory. In spite being closely located to testing spots, radiological situation at the most of conditionally «background» territories mainly depends on global fallouts. Except for zones of «plumes» of local fallouts from the first surface thermonuclear test with the yield of 400 kt (12.08.1953) and surface nuclear test with the yield of 38 kt (24.09.1951), made at the «Experimental Field» site. Fallout «Plumes» cross «southeastern» and «southern» parts of the STS under research and can determine different character of radioactive contamination of soil cover.

Investigation of radionuclides speciation was made using sequential extraction method with the following extragents applied: 1M  $\text{CH}_3\text{COONH}_4$  (pH=6,8) (exchangeable forms), 0,1n NaOH (bound to organic matter), 1n HCl (mobile or acid-soluble forms), widely used in worldwide science (Pavlotskaya F.I. 1974; Tessier A. et al, 1979).

As a result of performed researches low mobility of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  was found in soils notwithstanding type of fallouts, conditioned radioactive contamination of soils (Figure 1). The main concentration of  $^{241}\text{Am}$  transuranium radionuclide in soils was found in tightly bound form that is not accessible for plants, less significant concentration was noticed in mobile (acid-soluble) form, representing potential reserve for plants. As it was noted,  $^{90}\text{Sr}$  radionuclide is the most soluble in soils. Along with tightly bound forms of  $^{90}\text{Sr}$ , presence of easily accessible forms (exchangeable form) and forms representing potential reserve for plants (mobile form) was also found in soil. Herewith, the ratio between exchangeable and non-exchangeable forms of radionuclide is different, both for background sites and for the zones with increased radiation background. So

the result of investigation of  $^{90}\text{Sr}$  radionuclide mobility can be used as an additional parameter when studying the character of radioactive contamination of soil cover and when determining the impact of fallouts from surface nuclear tests on the territories under research.

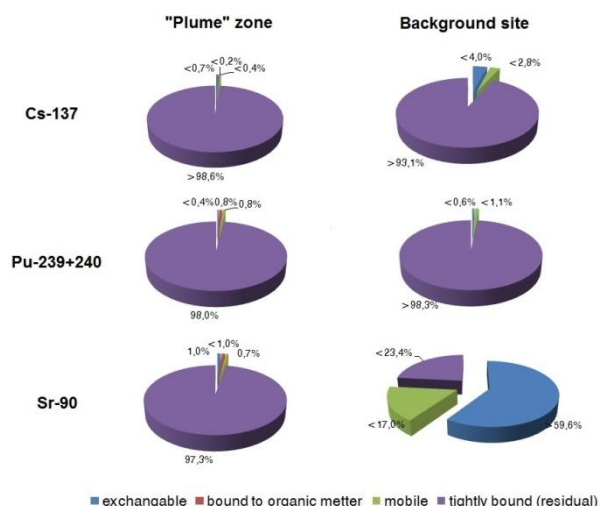


Figure 1. The species of radionuclides in soils of conditionally "background" territories of STS.

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# SOIL RADIOACTIVITY LEVELS AND RADIOLOGICAL RISK EVALUATION ACROSS THE STATE OF KUWAIT

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**Keywords:** naturally occurring radioactive materials, isotopic activity ratio, dose rates, hazard indices

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An evaluation of the surface radioactivity measurement associated with both naturally occurring radioactive materials and anthropogenic radio nuclei has been undertaken as part of a systematic study to provide a surface radiological map of state of Kuwait. The use of approximately 300 tons of depleted uranium shells during the First Gulf War has led to questions as to whether this material has a significant impact of the NORM levels in the surrounding environment [1].

Samples have been collected from approximately 185 independent locations across the state of Kuwait. These have been prepared and placed into sealed, marinelli beakers for a full gamma-ray spectrometric analysis using the high-resolution, low-background, high-purity germanium detection systems at the Environmental Radioactivity Laboratory of the University of Surrey using the procedure outlines in reference [2]. Of particular interest are the calculation of the activity concentrations associated with members of the decay chains following decays of the primordial radionuclides <sup>238</sup>U (<sup>226</sup>Ra, <sup>214</sup>Pb, <sup>214</sup>Bi) and <sup>232</sup>Th (<sup>228</sup>Ac, <sup>212</sup>Pb, <sup>208</sup>Tl).

This analysis includes evaluations for the <sup>235</sup>U decay chain. In particular, the 186 keV doublet transition is used together with the activity concentration values established from the decays of <sup>214</sup>Bi and <sup>214</sup>Pb to establish the <sup>226</sup>Ra and <sup>235</sup>U specific activity concentrations, which be used to estimate the <sup>235</sup>U : <sup>238</sup>U isotopic ratios compared to the expected expected for naturally occurring material of 1 : 138 [2]. Specific activity concentration values have also been determined for the <sup>40</sup>K and the anthropogenic radionuclides <sup>137</sup>Cs (from fallout) and <sup>134</sup>Cs (from the Fukushima accident) within the same samples. The paper will present an overview summary of the experimental samples which have been analyzed, including new data from different regions of Kuwaiti.

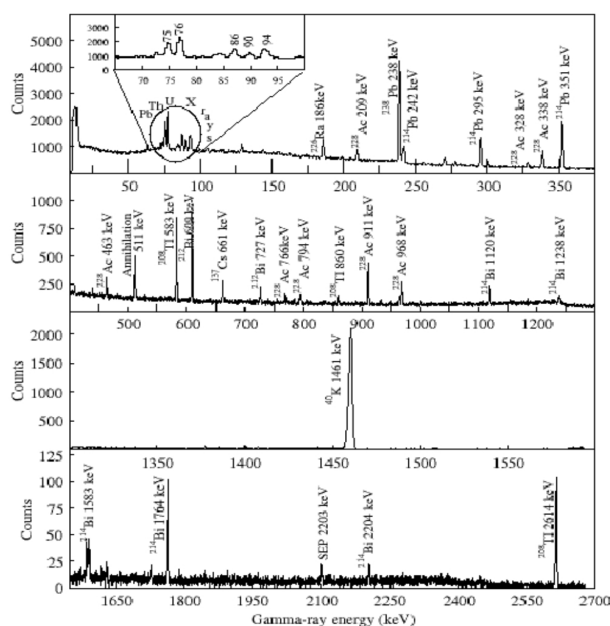


Figure 1. Measured soil spectrum showing gamma-ray transition from the <sup>238</sup>U and <sup>232</sup>Th decay chains.

## Acknowledgments

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## AIR MONITORING IN MONACO: STUDY OF SAHARAN DUST TRANSPORT

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Atmospheric input is continuously monitored and studied in the Principality of Monaco by IAEA-EL (International Atomic Energy Agency-Environment Laboratories) (Pham et al, 2011, 2012, 2013). Saharan dust events in the Northwest Mediterranean have been identified as important pathways for particle delivery into surface seawater (Moulin et al., 1997). The atmospheric transport of particles from North Africa to Monaco was recently studied at IAEA-EL (Pham et al, 2003, 2005). The study of radionuclides (both natural and anthropogenic) as well as the elementary composition of Saharan dust particles was carried out for better understanding the temporal behaviour of radionuclides in the atmosphere and their transport into the Northwest Mediterranean Sea.

Out of nine significant events registered episodically during 1998-2013, two important Saharan dust deposition events were detected at the Monaco air monitoring station of the IAEA during 23-24 November 2002 and 20 February 2004 (Pham et al, 2013), when a significant quantity of red-colour particles was collected. The influence of the 20 February 2004 deposition was further followed into the water column with the analysis of sediment trap samples collected at two depths - 200 m and 1000 m water depth at the Dyfamed station located at 43°25'N, 07°52'E (time-series studied from 21 December 2003 to 9 May 2004) (Miquel et al, 2011). The characterization of those particles was done using X-ray fluorescence (Spectro X-Lab 2000), gamma spectrometry (HPGe well-type detectors of 150% and 200% relative efficiency, Canberra model CW 15025-7915-30-ULB) operating in the IAEA-EL's underground laboratory with very low background, ICP-MS (Inductively Coupled Plasma-Mass Spectrometry) and AMS (Accelerator Mass Spectrometry) of Universidad de Sevilla, Spain. Data on the concentrations and activity ratios of natural and anthropogenic radionuclides (gamma emitters: <sup>7</sup>Be, <sup>40</sup>K, <sup>210</sup>Pb and <sup>137</sup>Cs; alpha emitters: <sup>210</sup>Po, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>239+240</sup>Pu; and Uranium isotopes) as well as major and trace elements are presented and discussed.

### Acknowledgments

The authors are indebted to many colleagues who contributed with sample collection. The IAEA is grateful for the support provided to its Environment Laboratories by the Government of the Principality of Monaco.

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## Deposition of radionuclides by cloud water on plants

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**Keywords:** cloud, radionuclides, deposition.

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After a nuclear accident like Fukushima, large quantities of radionuclides attached to particles are released in the atmosphere. Those particles can act as cloud condensation nuclei. In mountainous areas “occult” depositions such as cloud and fog water depositions can represent an important part of the wet deposition process. Therefore to determine the radiological impact of cloud water deposition on plants, an analysis of the cloud water radioactivity levels and a quantification of the cloud water deposition will be performed on the atmospheric research platform at the summit of the Puy de Dôme Mountain (1465 m a.s.l) in France. Moreover another goal of this study is to determine cloud deposition velocity to improve models and make them prone to accurately reproduce all kinds of wet deposition.

To collect cloud water samples, a replica of the Caltech Active Strand Cloudwater Collector will be implemented. This instrument allows air containing cloud droplets to be drawn through a cylinder by a fan. In this tube, rows of Teflon strands are collecting the droplets by impaction. Other active collectors have been developed on the same principle with a tube and a mesh grid as the impaction surface. Once the cloud water will be collected, it will be analyzed to determine the activity levels of gamma emitter radionuclides. In previous study (Bourcier *et al.*, 2014) the mean level activity for cesium 137, beryllium 7 and lead 210 in cloud water at the Puy de Dôme have been estimated. A comparison between activity levels found in rainwater and in cloud water highlights the fact that cloud deposition of radionuclides is far from negligible. At the same time a fog monitor FM-120 from DMT will provide the size distribution of droplets from 1 to 40 microns. A visibilimeter and a PVM from Gerber Scientific Inc. will provide respectively the visibility and the liquid water content (LWC) to identify and characterize cloud events.

In order to quantify the deposition of cloud water on plants, plastics plants will be exposed to cloud droplets deposition and weighed at the end of the event to measure the amount of water deposited. A second experiment will be conducted, during which plastics plants will be implemented on a polystyrene support; the whole set will be placed on a precision balance and under a protection box to avoid wind induced variations. The box will be removed for ten minutes for the cloud droplets to be deposited, then the box will be put back for weighing. Simultaneously another precision balance will be used to determine the deposition of water only on the polystyrene support in order to remove its weight from that of the previous set. A mass of water deposited by surface or mass unit of plant will be measured for each cloud event. Based on those flow measurements of cloud droplets deposition on plants and on the activity levels of collected cloud water, we will be able to determine values of cloud deposition velocity according to wind speed.

The results of cloud water activity levels and cloud deposition plants will be presented along with the first data of cloud deposition velocity related to wind speed.

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## Detection of Cs-137 in the Andes Cordillera at latitude 33°56' South.

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**Keywords:** Cs-137, Gamma Spectrometry, Soil, Andes cordillera.

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The anthropogenic radionuclide Cs-137 has been detected in different locations in the Southern hemisphere. It is a fission product and its presence in that part of the world is mainly due to the nuclear tests performed in the atmosphere in the Pacific atolls between 1966 and 1974. The geographical location of Chile, just in front of the Pacific Ocean, extending from 17° latitude South to Cape Horn at 56°, along the Andes cordillera, favoured the reception of fallout debris from those nuclear explosions occurred around 21.8° South. Even though the major precipitation of fission fragments took place years ago, long living radionuclides can be detected in soils.

The concern about the amount of environmental radioactivity to which the population is continuously exposed is stimulating new research in this field. We are interested in evaluating the contribution of these radionuclides to the radiation background in different zones of the country. Our preliminary studies on Cs-137 radioactivity in soil samples collected in the Andes mountains showed that after almost two half life, since those explosions, this radionuclide is easily detected (Correa, 2012). A cooperative research project between the University of Chile (UCH), the Metropolitan Technological University (UTEM), and the Chilean Nuclear Energy Commission (CCHEN), has been initiated. In a first stage concentrations of Cs-137 and natural radionuclides, in surface soil samples collected from the Andes to the Pacific Ocean will be determined. The first place where samples were collected is located at the Andes, specifically at Fundo Cruz de Piedra S 33° 56' 35.7" (latitude) and W 70° 11' 13.2" (longitude).

At the UCH laboratory the gamma detector is an ORTEC HPGe (GEM-10195) with resolution of 1,95 keV at 1330 keV and a relative efficiency of 10% reported by the manufacturer. The detector is shielded by a cylinder of lead, 10 cm thickness with internal covers of cadmium and copper, 1 mm thickness each. Spectra are being analysed by the code ROOT-Cern. Detector efficiency curve was obtained by using Certified Reference Material IAEA 447 (Shakhashiro, 2012). The efficiency of the gamma spectroscopic systems used in these measurements has been submitted to an internal intercomparison procedure between UCH and Environmental Radiation Section at CCHEN. In addition, the performance of the gamma analysis system at UCH was determined during ALMERA Proficiency Test (2012). Concentrations of radionuclide determined by this system showed differences of 5.5 % in Cs-137, 7.5 % in K-40, 8.3 % in Ac-228, 14.7 % in Pb-212 and 15 % in Tl-208 compared to values reported by IAEA.

Typical spectra corresponding to two soil samples collected in the Andes mountain are shown in Figure 1. Preliminary results point out that a significant peak to background ratio for Cs-137 is observed, indicating that concentrations are greater than the detection limit. A complete analysis of gamma spectra collected so far is being performed in order to determine activity concentration levels and associated doses from Th-232, K-40 and Cs-137.

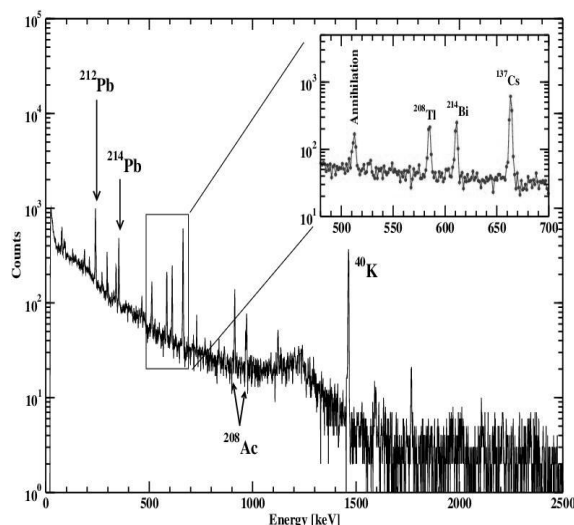


Figure 1. Gamma spectra collected from Chilean Andes cordillera. The inset shows the presence of anthropogenic radionuclide <sup>137</sup>Cs.

### Acknowledgments

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# Seasonality of $^7\text{Be}$ concentrations in Europe and influence of tropopause height

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Keywords:  $^7\text{Be}$ , interannual and seasonal variability, tropopause height, Europe

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Even though many studies have described surface  $^7\text{Be}$  concentrations at local sampling stations in Europe, few studies have performed a comprehensive and detailed analysis of its seasonal pattern at European level. More than improving the knowledge of its global distribution, the benefits of characterizing its temporal and spatial distribution might contribute to study changes in atmospheric processes (such as dry and wet deposition), to make easier the validation of global circulation models.

Therefore, the first aim of this work is to present a detailed description of spatial variability of the interannual and seasonal patterns of  $^7\text{Be}$  in EU. With this purpose, data collected from 2001 to 2010 by the Radioactivity Environmental Monitoring (REM) database in 17 representative locations have been analysed. The possibility to cover a relatively long time period (10 years), the use of a large number of sampling stations and the large data availability in this period increases the representativeness and validity of the results.

This analysis displays a latitudinal impact on the annual concentrations, with a tendency to register the lowest values in northern areas and the highest in southern regions. The analysis of interannual variability (Figure 1) reveals a general increase in  $^7\text{Be}$  activity concentrations in Europe that is associated with the decrease in the number of sunspots during this period (the end of the 23<sup>rd</sup> and the beginning of 24<sup>th</sup> solar cycles).

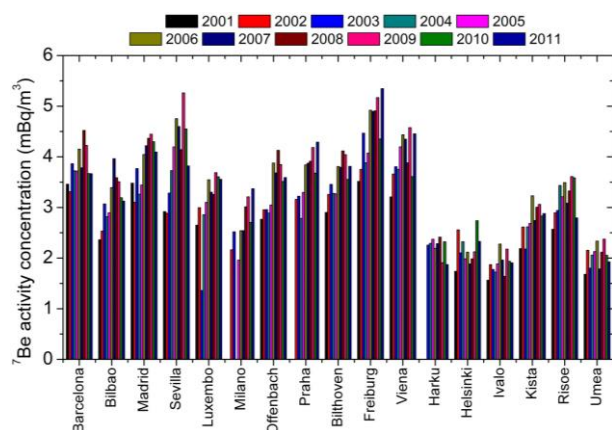


Figure 1. Interannual variation of  $^7\text{Be}$  in 2001-2010 period

Recent studies have reported the positive impact of the tropopause height as indicator for transport time of air masses enriched in  $^7\text{Be}$  to the surface. Those studies indicated a positive correlation in between the two parameters, explained by both downward transports of dry upper tropospheric air within anticyclonic conditions and lower scavenging rates. In addition, a time delay of 3 days has been found between the elevation of tropopause and  $^7\text{Be}$  concentrations in surface air.

Considering these previous results, the second aim of this study is to characterize the impact that the temporal evolution of the tropopause height has on  $^7\text{Be}$  activity concentrations and to infer the spatial distribution of the time lag between both parameters at European level. To this scope, we have determined the daily evolution of the tropopause height in each  $^7\text{Be}$  sampling station from 2001 to 2010.

The combined analysis of  $^7\text{Be}$  and tropopause height in each station displays a large spatial variability of the tropopause height impact on  $^7\text{Be}$  activity concentrations. The correlation coefficient between mean monthly atmospheric concentration of  $^7\text{Be}$  and tropopause height presents its maximum value in Madrid and Barcelona (southern area) (0.6-0.7), while the lowest values are obtained in Ivalo and Umea (northern area) (less than 0.2). The time lag between tropopause height and  $^7\text{Be}$  surface concentrations also shows a significant variation in Europe.

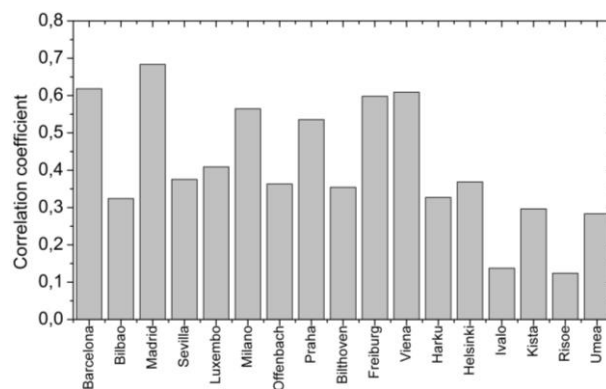


Figure 2. Correlation coefficient between mean monthly atmospheric concentration of  $^7\text{Be}$  and tropopause height (km) in 2001-2010 period

# Feasibility study on automatic evaluation of anomalous radioactivity levels using INTAMAP interpolation service within EURDEP network

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## Background

EURDEP (EUropean Radiological Data Exchange Platform) is both a standard format for radiological data and a network for the automatic exchange of unvalidated radiological monitoring data within Europe, in both routine and emergency mode. (<https://eurdep.jrc.ec.europa.eu>).

A continuous exchange of information in nearly real-time of environmental radioactivity levels (ambient dose equivalent rate, radioisotope air concentration) as well as meteorological parameters (pressure, temperature, wind direction and speed) collected at measuring stations placed in 37 European countries is performed through the EURDEP network. The data exchange is mostly done on hourly basis; both during routine and emergency operation, but the frequency could be increased in case of emergency. To visualize this large amount of data, EURDEP provides, among other tools, a web-based GIS (Geographical Information System) map (<https://remap.jrc.ec.europa.eu/>).

Born out of a need of continuously improving the EURDEP network, and always with the aim of facilitating its use and understanding by the competent authorities, European Commission works for implementing new specialized tools for improving the interpretation and understanding of the monitoring results in both routine and emergency situations.

Within this context, the interpolation of Radiological environmental based on the data provided by the national environmental monitoring networks plays a key role, especially in emergency situations. A feasibility study to evaluate the possibility of implementing a function based on the results of the Interoperability and Automated Mapping (INTAMAP) project (<http://www.intamap.org/>) for generating automatic spatial interpolated data in EURDEP has been triggered.

## Objectives

One of the first points to be addressed in a radiological/nuclear early warning system is the verification/evaluation of the high measurements which are automatically registered by the sampling and measuring stations. In this kind of events, the values can be 1) higher than the threshold level or 2) close to the threshold value. These two scenarios can cause either false positives or false negatives, and therefore, the validation of the values needs to be addressed to guarantee the proper functioning of the Early Warning System.

The feasibility study presented in this work is aiming at facilitating the automatic detection and evaluation of anomalous values, which could be later confirmed as real high values, by implementing INTAMAP automated interpolation service imbedded in the EURDEP platform.

## Methodology

Such implementation exploits the ability of INTAMAP automated interpolation service to deal with data containing extreme outliers using the copula kriging, which handles well the extreme values and provides the estimation of the probability that a critical value was exceeded or not. However, it is necessary to consider the fact that this computational technique is quite time-consuming.

The main purpose of this interpolation request is to look at the probability of fitting the registered unusual value under the copula predictions. So that if the real measurement result remains outside a reasonable range of quantiles [e.g. 0.01 - 0.99] the system automatically warns against a possible anomalous event. The step by step methodology is explained in Figure 1.

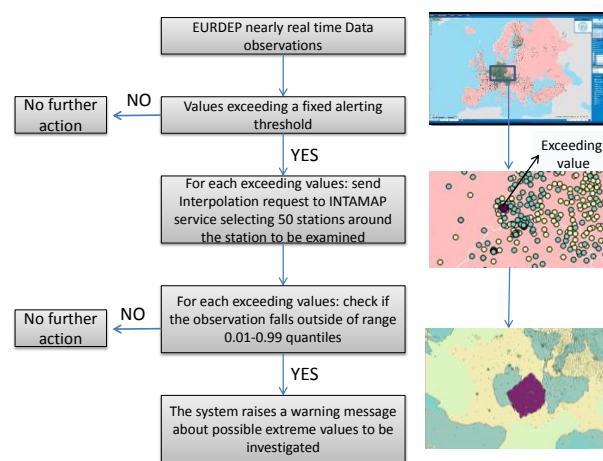


Figure 1 – step by step methodology.

## Results

The method implemented will be tested against several scenarios also considering real past cases, in order to set a benchmarking useful for better improving its usage. The results obtained have presented a good agreement, proving the validity of the methodology, although more sensitivity studies and further research need to be addressed.

# Beryllium-7 and $^{210}\text{Pb}$ atmospheric deposition measured by a moss technique and its dependence on cumulative precipitation

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**Keywords:**  $^7\text{Be}$ ,  $^{210}\text{Pb}$ , atmospheric deposition, cumulative precipitation.

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Natural airborne radionuclides  $^7\text{Be}$  and  $^{210}\text{Pb}$  are frequently used as tracers in atmospheric studies. Once after formed,  $^7\text{Be}$  and  $^{210}\text{Pb}$  due to high chemical reactivity are attached to aerosol particle and in this form follows all atmospheric paths of transport and deposition of aerosols. The measurement of  $^7\text{Be}$  and  $^{210}\text{Pb}$  activity concentration (in  $\text{Bq m}^{-3}$ ) in atmosphere is usually done by air sampling or by deposition samplers to determine activity radionuclide in fallout (in  $\text{Bq m}^{-2}$ ). Temporal variations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  contents in air and precipitation were a subject under consideration numerous studies (Ioannidou et al., 2005) and in some cases correlation between measured airborne radionuclide and some atmospheric parameters (precipitation) was also subject of research.

Currently, terrestrial mosses are widely used in environmental monitoring. The moss technique has shown to be very suitable for studying the atmospheric deposition of heavy metals as well as other trace elements, including radionuclides.  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities measured in mosses are not correlated with precipitation amount mostly for the reason that that mosses cumulate activity (Krmar et al., 2009). It means that some different approach in analysis of relations between precipitation and moss activity should be considered. The most important goal of this study is to explore some techniques which can be able to give more insight in possible relationships between measured activities of airborne radionuclides in collected mosses and amount and duration of precipitation.

Samples of naturally growing mosses were collected from single location one sample per week, during 14 months. Dry moss samples were packed in standard geometry and measured by 100% relative efficiency low-background HPGe detector equipped with a lead shielded (16 cm) and carbon window. The daily amount of precipitation was taken from the nearest meteorological station that is in the network of meteorological stations of Hydro-meteorological Institute of the Republic of Serbia.

We used data of cumulative precipitation and cumulative duration of precipitation and derived "cumulative activities" (calculated by simple addition of successive measured values until the end of the studied period of time) and "running activities" (decay – corrected sum of  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities in mosses) to explore relations between those datasets. Considering that possible variation of  $^{210}\text{Pb}$  and  $^7\text{Be}$  in measured samples of mosses could be a result of some local variations the relationship between cumulative activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  should be independent of some similar

factors. To involve possible influence of precipitation or duration of precipitation, the measured cumulative activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  were normalized by cumulative precipitation and cumulative duration of precipitation (Fig. 1).

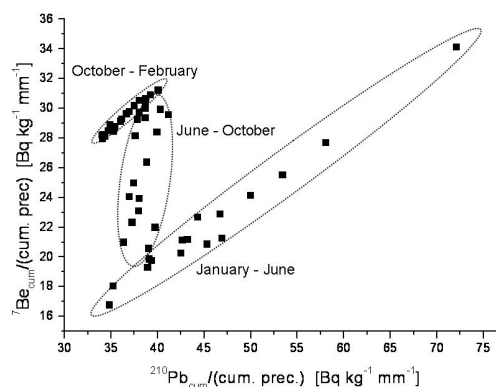


Figure 1. Correlation between cumulative activity of  $^7\text{Be}$  and  $^{210}\text{Pb}$  normalized by the cumulative precipitation

Points on scatter graphs are grouped in clusters. Values obtained in the period January – June are grouped in one cluster indicating constant ratio between them. Data from June until October are showing significantly different ratio between normalized  $^7\text{Be}$  and  $^{210}\text{Pb}$  cumulative activities. It is period of the increase of  $^7\text{Be}$  deposition which gives more intensive increasing trend of cumulative activity concentration of  $^7\text{Be}$  comparing with obtained values of  $^{210}\text{Pb}$  cumulative activity. Data obtained in the period between October and February are grouped in third cluster. It can be seen that ratio between normalized cumulative  $^7\text{Be}$  and  $^{210}\text{Pb}$  in first and third group of data are similar.

## Acknowledgments

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# Applications of environmental radionuclides to atmospheric studies at the WMO-GAW station of Mt. Cimone

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**Keywords:**  $^7\text{Be}$ ,  $^{210}\text{Pb}$ , stratosphere-troposphere exchange, back-trajectory, chemistry and transport model.

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Radionuclides have been traditionally employed in environmental studies as "tracers" of environmental processes, providing timescales of processes and trace substances involved in terrestrial transport, exchange and mixing processes (Papastefanou, 2008; Froehlich and Masarik, 2010). In the atmospheric sciences, the research on cosmogenic and terrigenous radionuclides has been crucial for a better understanding of several basic processes, such as interhemispheric transport (Froehlich and Masarik, 2010), stratosphere-troposphere exchange (STE) (Reiter, 1971; Zanis *et al.*, 2003) and timescales of atmospheric dynamics (Huh *et al.*, 2006). Moreover, it also provided important insights into transport and/or deposition processes of atmospheric pollutants (e.g., ozone and particulate matter).

In this work we present an overview of our research activity on airborne radiotracers concerning the long term observations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in  $\text{PM}_{10}$  matrix collected over the past twelve years at the WMO-GAW (World Meteorological Organization-Global Atmosphere Watch) station of Mt. Cimone (44.20 N, 10.70 E; 2165 m a.s.l.; Italy) (Lee *et al.*, 2004; Tositti *et al.*, 2014). Owing to their contrasting origins, the pair of natural radionuclides  $^7\text{Be}$  and  $^{210}\text{Pb}$  ( $^7\text{Be}$  is a cosmogenic radionuclide, while  $^{210}\text{Pb}$  is a radionuclide of crustal origin from  $^{222}\text{Rn}$  decay) are among the most used naturally occurring radionuclides. The results presented include both innovative and classical studies such as the use of  $^7\text{Be}$  in the identification of stratosphere-to-troposphere transport, which inspired the initial research activity in this field.

In the former case the results we obtained showed that differently from what previously believed mid-latitudes STE events are not uniquely typical of the transition between winter and spring, but they occur all over the year (Bonasoni *et al.*, 1999, 2000a,b; Cristofanelli *et al.*, 2003, 2006, 2009). While these researches inherently suggested the importance of the use of the ratio  $^7\text{Be}/^{210}\text{Pb}$  in the study of vertical motion in the troposphere, the need for more quantitative modelling analyses arose. This led to the applications presently in progress in our group, resulting from the collaborations with Dr. Miguel A. Hernández-Ceballos (JRC, Joint Research Centre of the European Commission, Ispra, Italy), Prof. José Antonio García Orza (Universidad Miguel Hernández de Elche, Spain) and Dr. Hongyu Liu (NASA Langley Research Center, Hampton, VA, USA). Our research objectives are: 1) characterize the

advection patterns at Mt. Cimone and their influences on atmospheric composition; 2) better understand the seasonal variations of atmospheric radiotracers as well as of those of the roles of transport and precipitation scavenging in their seasonal variation (Figure 1) (Brattich *et al.*, 2015; 4 papers in preparation).

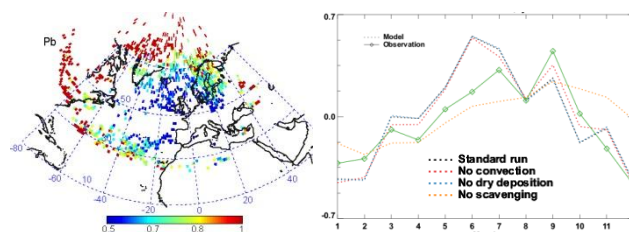


Figure 1. Examples of application of statistical analyses on back-trajectories together with  $^7\text{Be}/^{210}\text{Pb}$  ratio (left) and applications of the Global Modeling Initiative (GMI) chemistry and transport model (CTM) to elucidate the roles of different processes on  $^{210}\text{Pb}$  seasonal variations at Mt. Cimone (right).

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# Radionuclide deposition with snow on Mt. Zugspitze, Germany: Washout and release to surface water

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**Keywords:** radioecology, snow, washout, radionuclide deposition, surface water

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Snow scavenging is supposed to be the most efficient process in the removal of aerosol particle bound radionuclides from the atmosphere. But their transfer into the snow cover and release to snowmelt are sparsely investigated. However, they are important due to a) high efficiency of radionuclide scavenging by snow, b) large spatial distribution and long term accumulation of snow, c) concentration by sublimation and partial melting, d) delayed release of radionuclides from the snowpack in winter and short term release to surface water in spring, and e) climate change with increase of the probability of extreme weather phenomena.

The experimental work was carried out at the Environmental Research Station Schneefernerhaus on Mt. Zugspitze, Germany (2,650 m a.s.l.) using Be-7, Pb-210 and Cs-137 as trace radionuclides. Snow characterisation was carried out with a 2D video disdrometer combined with a precipitation gauge, delivering parameters of snowfall intensity, type of precipitation, snow crystal size, velocity and shape distributions as well as number concentrations. Particle scavenging in the size range of 10–550 nm was quantified by SMPS (scanning mobility particle sizer). From continuous 10 min interval measurements, scavenging coefficients for washout of aerosols through precipitation were calculated. Snow height, snow water equivalent, densities and liquid water contents of the snowpack were measured by a snow pack analyser and a snow scale. Radionuclide deposition was determined by gamma-spectrometry in monthly snow samples.

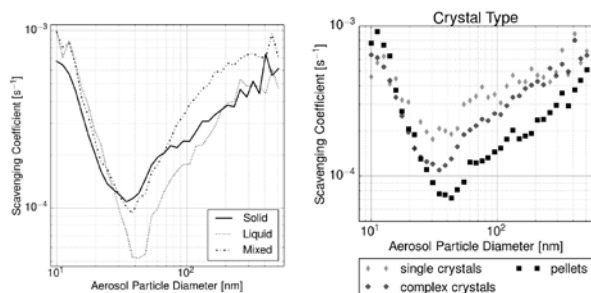


Figure 1. Calculated scavenging coefficients for a) rain, mixed and snow events and b) different types of snow crystals related to the aerosol particle diameter.

Mean scavenging coefficients were calculated with  $(3.2 \pm 0.8) \cdot 10^{-4}$  for rain,  $(3.3 \pm 0.8) \cdot 10^{-4}$  for snow and  $(4.5 \pm 1.0) \cdot 10^{-4}$  for mixed precipitation events. The highest scavenging coefficients were calculated for mixed events of snow and rain for particle sizes > 30 nm due to the existence of large snowflakes with high water contents that fall slowly (Fig. 1a). The higher the precipitation rate the stronger is the scavenging for liquid and for solid precipitation. Related to the shape of snow crystals single crystals (e.g. dendrites) with large surface areas are the most efficient scavengers for small aerosol particles. Rounded crystals like graupel and pellets have the smallest washout coefficients (Fig. 1b). Therefore, the measurement of snow crystal characteristics is necessary for the prediction of radionuclide washout by snow.

Once deposited in the snowpack the radionuclides accumulate until first snowmelt is initiated. Deposited amounts of Be-7, Pb-210 and Cs-137 were determined in monthly snow samples since beginning of the snow season 2014/2015 in October 2014. At the beginning of the snowmelt period, snow height decreases due to compaction of the snowpack. The liquid water content stays constant, but water saturation of the lowermost snow layer begins. At a certain point, melt water is released, which can be a fast process. With the first 20 % of melt water > 80 % of the radionuclide content is released. In May 2014 the release happened in two steps. First, 12 % of the melt water was set free in 7.5 d. After a following freezing period, again 20 % of meltwater were released in 6.3 d. This means that at step 1 already about 50 % of the radionuclide inventory of the snowpack were released. At step 2 after 6.3 d another 80 % of the remaining radioactivity were in the runoff.

In conclusion, snow may deposit radionuclides with a high efficiency which can be released in spring in a very short period of time and may contaminate surface and drinking water reservoirs directly.

## Acknowledgements

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# SEASONAL VARIATIONS OF RADIOACTIVITY IN AEROSOLS AT ALGIERS STATION, ALGERIA

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Keywords: Seasonal variations, Radionuclides, gamma spectrometry and Mean residence time

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The great variation of air activity concentrations of natural short-lived radionuclides in atmospheric air depends on the place, time, height above the ground and weather conditions. Moreover, it is influenced by the transport in the atmosphere with the associated aerosol particles, and by radioactive decay. The removal processes from the atmosphere have a dominant influence on the activity concentration of long-lived radionuclides (Ahmed et al., 2004).

The residence time of atmospheric aerosol particles in the lower atmosphere, assuming that the troposphere is considered a well-mixed reservoir (closed system), is a function of various removal processes, the most important being:

- (a) dry deposition by impaction, diffusion and sedimentation, and;
- (b) wet deposition by rain drops (precipitation scavenging) (Papastefanou, 2008).

The residence time of atmospheric aerosol particles can be estimated by means of radioactive nuclides as tracers, which become attached to aerosol particles and are removed with them as they are scavenged by precipitation or undergo dry fallout (Warneck, 1988).

The main purpose of this work is to understand transfer of natural radionuclides in the atmosphere, such as the cosmogenic radioelement <sup>7</sup>Be and some decay product of uranium series, mainly <sup>210</sup>Pb, and to estimate the mean residence time. It is also helpful to detect any other input of artificial radioactivity.

During a one year period, from October 2011 up to October 2012, approximately, 41 air filters were sampled and analyzed to determine the atmospheric air activity concentrations of cosmogenic radionuclide (<sup>7</sup>Be), and long lived (<sup>222</sup>Rn) decay product <sup>210</sup>Pb, and an eventual artificial radioactivity.

With a flow rate of 67.9 m<sup>3</sup>.h<sup>-1</sup>, the air was sampled through paper filters. The sampling station is located at an altitude of 157 m above sea level at Nuclear Research Center of Algiers.

Filters were analyzed in the laboratory by direct counting by gamma spectrometry using high purity coaxial germanium detector with a 40% relative efficiency.

Most of radioactivity measured during the period of this study is natural.

The monthly concentrations of <sup>7</sup>Be show clear seasonal changes with high values during spring-summer and low values in autumn-winter.

Highest values of the mean monthly concentrations of <sup>210</sup>Pb were observed in summer and the lowest in winter.

Table1. Seasonal <sup>7</sup>Be and <sup>210</sup>Pb concentrations during the investigation period.

Season	<sup>210</sup> Pb (μBq/m <sup>3</sup> )	<sup>7</sup> Be (mBq/m <sup>3</sup> )
Autumn	333.1±16.7	4.81±0.24
Winter	197.13±10.72	4.43±0.22
Spring	320.25±16.82	5.34±0.27
Summer	570.11±27.25	5.2±0.26
Mean	333.27±17	4.91±0.25

Correlations between the atmospheric radioactivity (<sup>210</sup>Pb and <sup>7</sup>Be) and meteorological parameters (air temperature, precipitations and relative humidity) were determined.

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# Influence of iodine supply on the radiation-induced DNA-fragmentation

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**Keywords:** radioiodine, environmental radioactivity, radiation sensitivity.

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Any unwanted release of radioiodine to the environment may cause an increased risk of thyroid cancer. This was clearly demonstrated after the Chernobyl accident in 1986 that led to a statistically significant elevation of thyroid cancer under the exposed population [Shakhtarin et al. (2003)] and was especially observed for exposed children. Iodine deficiency in the respective population aggravates this risk substantially. The reduction of this cancer risk is the rationale for administration of stable iodine in form of potassium iodide after any accidental exposure to radioiodine. Up to now the justification of this procedure is the assumption of reduced uptake of radioiodine due to the excess of stable iodine that saturates the transport mechanisms into the thyroid cells.

The uptake of iodine into thyroid cells is regulated by the NaI-symporter (NIS). The radioiodine ("hot iodine") uptake in cells is limited by the presence of stable iodine ("cold iodine") due to the competition for the NaI-symporter ("competition effect") of these two isotopes of the same element.

Stable iodine itself gives rise to cellular effects that might influence the radiation sensitivity irrespective of the above-mentioned competition effect. Therefore, this work focusses the question of a possible radioprotective role of iodine regardless of the competition in terms of cellular uptake.

Thyroid cells (FRTL-5) were externally irradiated by use of a linear accelerator (LINAC) and by incubation with various concentrations of <sup>131</sup>I. Skin cells (no NIS) were equally treated in order to investigate the distinctiveness of thyroid cells. Doses between 0.01 Gy and 400 Gy were applied. For the different types of radiation exposure we investigated the influence of the concentration of stable iodine added to the cells prior to irradiation.

Radiation damage was determined by assessing of apoptosis-like intracellular DNA fragmentation or necrosis-like extracellular DNA fragmentation (CDDT, cell death detection test). The extent of radiation damage was expressed as enrichment factor. DNA fragmentation is one of the most important effects of ionizing radiation in cells.

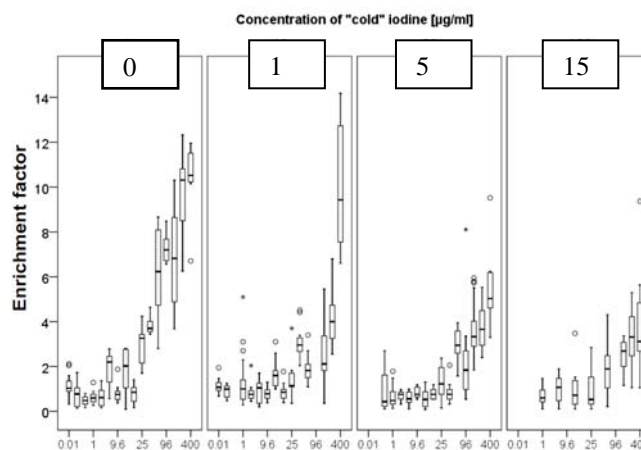


Fig. 1: Dose-dependent radiation-induced DNA damage after addition of amounts of stable iodine

The addition of "cold" iodine (1 to 15 µg/ml) without irradiation ("negative control") did not change the DNA fragmentation for both cell types. The DNA fragmentation of both cell types did increase markedly with increasing radiation dose. This radiation effect is diminished if iodine is added to the thyroid cells beforehand (Fig. 1). The skin cells, however, did not show a significant radiation induced change of DNA fragmentation in presence of cold iodine. It may be concluded that elementary iodine significantly reduces DNA-fragmentation. This effect of iodine is seen only in cells with NIS.

Obviously, there are two independent biological mechanisms with a radioprotective effect on thyroid cells. Besides the well-known prevention of radioiodine uptake stable iodine has a direct effect on the radiation sensitivity of thyroid cells.

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## Cs-137 in oak bark samples from Romania and Serbia

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The Chernobyl disaster from April 26/27, 1986 deposited variable quantities of Caesium-137 and other radioisotopes over the whole Europe. There is high scientific interest to study the <sup>137</sup>Cs content in forest ecosystems due to the fact that forests are complex environments with great capacity to intercept and to retain radionuclide deposition and other pollutants for a long time [1].

The bark contamination was dependent of roughness and absolute bark surface, also of leaves/needles presence at the beginning of May 1986. In the forest ecosystems, nearly 75-80% of the Chernobyl deposition remained in the upper part of the canopy in the first period (~ 2 months) following the radioactive release. In the following 3 years 80% of <sup>137</sup>Cs from the canopy reached soil surface by litter-fall, through-fall and stem-fall. A part of <sup>137</sup>Cs from leaves (needles) was removed by raining or wind and was captured in the trunk bark [2-4]. In Romania, the higher Chernobyl deposition was at the beginning of May, 1987, on NE - SW direction and it is assumed that the radioactive cloud continued its trajectory in Serbia on the same axis, finally reaching the southern of Italy. Nowadays, <sup>137</sup>Cs can still be found in various quantities in the tree bark [5, 6]. As it was shown [7] bark of different tree species have various retention capacity for <sup>137</sup>Cs and oak bark proved to provide the highest <sup>137</sup>Cs concentrations.

In this work, bark samples of oak trees older than 60 years were considered, collected at 1.3 - 1.5 m height from the soil using a medium chisel. Powdered bark samples were measured by gamma spectrometry in cylindrical geometry, placed on the top of the vertical HPGe detector of relative efficiency 30%. The detection limit (95% significance level) was about 1.5 Bq kg<sup>-1</sup>. The measurement result uncertainties were determined in a range between 2–36% by applying the error propagation law.

During 2014-2015 period, more than 80 oak bark samples from three Serbian regions (the surroundings of Beograd, Niska Banja and Novi Pazar) and four Romanian regions (Cluj-Napoca area, Ciomad-Balvanos reservation, Hateg county and Herculane area). Preliminary results show a strong correlation

between the present content of Cesium-137 in oak bark samples and the presence of rainfalls in the first half of May, 1986.

### Acknowledgments

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## Progress on a Voxel Phantom Model of a Pine Tree

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Radiological protection has historically focused on the protection of humans, but there is now a focus on including the interactions between radionuclides and the environment. Research has been conducted on some of the International Commission of Radiological Protection (ICRP) reference animals that have resulted in voxel phantom models, but there are still no voxel phantom models for the ICRP reference plants.

ICRP 108 provides an ellipsoidal model of the pine tree that takes into account a cylindrical trunk and an ellipsoidal top with uniform distribution of radionuclides throughout the modeled mass. The model currently under development will have a more accurate measurement of living mass for dose calculations, as there is a vast difference between the living and dead mass of a pine tree. The outer layers of the pine tree trunk are alive and exchange nutrients and water. The inner layers of the pine tree are heart wood and are not living tissue. Similarly only parts of the root system are still living while others have seasonally dependent death and growth. This distinction in living mass and total mass of a pine tree will greatly change the mass used in dose calculations for pine trees.

These changes in understanding of the biology of a pine tree underscore the need for a more accurate model of a pine tree to improve the understanding of potential radiological effects and may shed light on the biological mechanisms that make pine trees so radiosensitive.

A greenhouse trial with pine tree seedlings provides information on the biological growth of pine trees. After a period of measurements some of the plants are slated to be disassembled and specific masses of the plant will be taken and the remaining plants will be used for full CT scans. CT scans are utilized to develop a voxel phantom model using 3D doctor. Monte Carlo N-Particle (MCNP) can then be utilized to calculate absorbed fractions and subsequently make dose estimates to specific tree organs.

These absorbed fractions will be used to calculate dose rates to environmental samples from radionuclides exposure sites to better understand pine tree radionuclide interactions.

This research procedure will provide a voxel phantom model which will help illuminate the biological responses of tree tissues to radiation exposure.

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## **SEMIPALATINSK TEST SITE (STS). PRESENT STATE AND PROSPECTS.**

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Voluntary closing of the STS in conditions of the USSR and its consequent collapse (in spite of existing projects on provision of international assistance) in fact have left Kazakhstan alone facing its problems related to rehabilitation of the territory and population of the region. Furthermore as necessity to provide nuclear and radiation safety for both research reactors located at the territory of the former STS, as well as places where nuclear tests were performed those contain nuclear waste and potentially dangerous for the non-proliferation criteria.

The test site is situated at crossing of three regions of Kazakhstan: East-Kazakhstan, Pavlodar and Karaganda and occupies an area of 18,5 thousands sq.km. During the period of the test site operation (1949-1989) 456 nuclear tests were conducted at its territory, including 30 land surface tests, 86 atmospheric tests and 340 underground nuclear explosions. By the Order of the President of the Republic of Kazakhstan of the 29<sup>th</sup> of August, 1991 No.409 STS was closed. Works on elimination of consequences of nuclear tests, started after the test site was closed, including elimination of infrastructure for testing of nuclear weapons, conversion of objects of military industrial complex, objective assessment of scale and levels of radioactive contamination of environment, as well as development and implementation of measures, preventing influence of nuclear tests consequences on population health. Complexity for formation of radiation situation is associated with different types of tests performed at the test site (nuclear tests (atmospheric tests, underground, excavation tests), hydronuclear tests, testing of combat radiological agents, and possibly other experiments we don't know), as well as climatic and natural conditions (storm winds, different geological and hydrological conditions), determining migration processes of radionuclides. The main sites of the STS used for the tests listed above are: «Experimental Field», «Degelen», «Balapan», «Sary-Uzen», «Telkem», "4a" and "4".

Three of four research nuclear reactors existing in Kazakhstan are located in the STS territory. They are located on the two experimental complexes of the National Nuclear Centre of the Republic of Kazakhstan. One of those also involves long-term storage place for spent encapsulated sources of ionizing radiation which has national significance. In their days these reactors were used to make different experiments, associated with significant emissions of radionuclides into environment. Radiation environment near these objects haven't been studied completely yet.

Since the test site was closed till nowadays NNC RK in cooperation with other specialists from Kazakhstan and the international scientific society obtained a large volume of information about present radiological state of the STS and adjacent territories. All significant radioactively contaminated territories, as well as main routes and mechanisms of current and potential distribution of radioactive substances were found. Obtained data allow concluding, that currently the STS have no negative impact on population, living in the adjacent territories, except for people living in the impact zone of Shagan river. Compliance with the law and specially developed rules regarding activities at the STS territory provides radiation safety when performing economic activities in the STS. Radioecological situation there is not stable, migration of radioactive substances were revealed and that make it necessary to provide regular monitoring of radiological situation in the STS.

Certainly, taking into account the scale of the STS and variety of tests made here, available information is not comprehensive, but it allows proposing a scientifically grounded plan of research works and practical liquidation (remediation) measures. These works should result in transfer of up to 90 % of the STS lands into economic turnover.

# Bioaccumulation of the natural radionuclides Th-234, Ra-226 and K-40 and the artificial Cs-137 in the fruitbodies of Basidiomycetes in Greece

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**Keywords:** Basidiomycetes; Natural Radionuclides; Cs-137; Bioindicators; Concentration Ratios  
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This study reports on the bioaccumulation of artificial Cs-137 and natural radionuclides Th-234, Ra-226 and K-40 by species of Basidiomycetes.

Thirty two composite samples of fungal fruitbodies (Basidiomata), representing thirty species of Basidiomycetes, were collected with their corresponding soil samples from six sampling areas of 5 - 20 m<sup>2</sup> in central and northern Greece, between September 2002 and November 2002.

The activity concentrations of the artificial Cs-137 and natural radionuclides Th-234, Ra-226 and K-40 for both the fungal and the soil samples were measured for 70000 sec in a high-resolution gamma spectrometry system with an high purity Ge-detector of 20% relative efficiency and computerized multichannel-analyzer of 4000 channels in a total spectrum area of 2000 keV, established in ERL/INRASTES/NCSR"D".

The results show that the activity concentrations for Cs-137 ranged from  $0.2 \pm 0.0$  to  $87.2 \pm 0.4$  Bq Kg<sup>-1</sup> Fresh Weight (F.W.), for Th-234 from  $1.5 \pm 0.9$  to  $30 \pm 25$  Bq Kg<sup>-1</sup> F.W., for Ra-226 from  $0.3 \pm 0.1$  to  $1.0 \pm 0.5$  Bq Kg<sup>-1</sup> F.W. and for K-40 from  $56 \pm 3$  to  $760 \pm 30$  Bq Kg<sup>-1</sup> F.W. (Figure 1).

By analyzing the data, it is supported that Bioaccumulation is dependent on the fungal species and only in the case of Cs-137 on the functional group of the Fungi, after applying the Kruskal Wallis Test (Table 1).

The Concentration Ratios (CRs) for each fungal species were also calculated, as a means of estimating the transfer of the radionuclides from soil to the fungal fruitbodies, thus assisting in selecting potential bioindicators for each radionuclide for the early impact assessment in the terrestrial environment, in cases of radiological events under routine or accidental releases.

In terms of the artificial Cs-137: a) *Ramaria formosa* collected in Agrafa presented the highest value ( $87.2 \pm 0.4$  Bq kg<sup>-1</sup> F.W.), even though its substratum soil did not present the respective highest one, b) subsequently, *Ramaria formosa* also presented the highest CR (0.28), with the lowest one observed in *Lycoperdon perlatum* (0.0005).

Concerning the natural radionuclides, a selective bioaccumulation of Th-234 was observed, instead of its parent U-238 in Fungi. Among the 18 species considered, *Sarcodon martioflavus* presented the highest value of activity concentration ( $30 \pm 25$  Bq kg<sup>-1</sup> F.W.) and a CR of 0.96. The lowest value for CR was found in *Boletus* sp. (0.043). For Ra-226, *Hydnellum concrescens* presented a CR over an order of magnitude higher than that of the other species (0.035). For K-40

the highest CR was found in *Sarcodon martioflavus* (1.50).

As general conclusions, it can be noted that: a) mycorrhizal Fungi were better indicators for the Cs-137 impact assessment than saprotrophic and parasitic ones, b) a ranking as:  $CR_{K-40} > CR_{Th-234} > CR_{Cs-137} > CR_{Ra-226}$ , was observed for all the radionuclides reported.

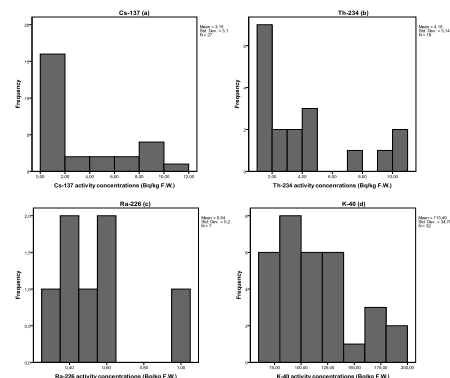


Figure 1. Distribution histograms of activity concentrations (Bq kg<sup>-1</sup> F.W.) for the radionuclides Cs-137 (a), Th-234 (b), Ra-226 (c) and K-40 (d).

Table 1. Kruskal Wallis Test results in the form of p-values for Cs-137, Th-234, Ra-226 and K-40

Kruskal Wallis Test Grouping Variable:	p-values			
	Cs-137	Th-234	Ra-226	K-40
Functional group	0.032	0.236	0.718	0.738
Family	0.298	0.317	0.826	0.462
Sampling site	0.358	0.522	0.667	0.237

## Acknowledgements

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# RADIONUCLIDES IN BODIES OF WILD ANIMALS OF SEMIPALATINSK TEST SITE

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During testing of nuclear weapons and adverse factors of radioactive substances at the territory of Semipalatinsk Test Site (STS) spots with high level of radionuclides in such natural environments as soil, vegetation and water were formed. As a rule, such territories are associated with epicenter of nuclear explosions at "Experimental Field" site, where surface nuclear tests were made, channels of radioactively-contaminated streamflows from "Degelen" testing site (Panitskiy, 2015), places of warfare radioactive agents testing at "4" and "4A" sites and places of underground excavation nuclear explosions – "Atomic lake", "Telkem 1", "Telkem 2" (Lukashenko, 2014). Radioactive contamination of the test site territory was also noticed in form of "plumes" resulted from passing radioactive clouds formed at the moment of tests.

Due to high enough concentrations of artificial radionuclides found in soil, vegetation and water, concentrations of *определенный интерес* artificial radionuclides in organisms of animals living in such areas in holes in radioactively contaminated soil is of some definite interest. That is why as a research object we selected reptiles and mouse-like rodents. A set of their biological peculiarities (including relatively long life of individuals, sedentary life, close association with substrate, low activity radius, high abundance, stability of main population characteristics, simple capturing and observation process) make them perspective for research. Moreover, these animals are the component of food chain for predator animals and birds.

Data given in this paper were obtained within the frame of ISTC international projects (K-759, K-2085 projects) and state republican programs of the Republic of Kazakhstan.

Sand lizard (*Lacerta agilis Linnaeus*) and stepperunner (*Eremias arguta pallas*) were selected as research objects. Both species are representatives of *Lacertidae* family widely spread at the STS territory. Biological peculiarities of both species are well studied.

Animals were captured at different parts of the test site, including "4A" site within the area of "Atomic" lake, at different parts of "Degelen" site and conditionally

clean ("background") territories of the STS beyond testing spots of the test site with concentrations of radionuclides in soil corresponding to background levels of global fallouts established for the northern hemisphere.

After capturing animals were delivered to the laboratory and narcotized with diethyl ether. Animals were affected by ether vapors till loss of vital signs. Bodies were flushed in influent water, and disintegrated after removal of internal organs. Muscular tissue of mouse-like rodents was separated from bone tissue.  $^{137}\text{Cs}$  was determined in well homogenized raw mass.  $^{90}\text{Sr}$  was determined in ash obtained as a result of drying the raw mass and ashing at the temperature of 500 °C to constant mass.

Performed researches show that concentrations of radionuclides in organisms of wild animals, living in different parts of the STS differ from one another. The main radionuclide contributing to contamination of lizards in ecosystems associated with radioactive stream flows is  $^{137}\text{Cs}$ . At WRA testing sites it's—  $^{90}\text{Sr}$ . At that concentrations of radionuclides in lizards' organisms depend on source levels of radionuclides concentration in components of live environment and forms of radionuclides occurrence in soil. In some definite conditions (like in case of WRA testing site) lizards can significantly contribute to redistribution of radionuclides in natural environment. So at some individual radioactively contaminate parts of this site very high specific activities of  $^{90}\text{Sr}$  radionuclide achieving value of  $7.8 \times 10^5$  Bq/kg were found in lizards' bodies.

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## Parameters of radionuclide transfer to livestock and poultry products in distant period after nuclear tests at the Semipalatinsk test site

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Since 2007 at the Institute of Radiation Safety and Ecology NNC conducted research of parameters of radionuclide transfer in livestock and poultry products. The focus is on radionuclides  $^3\text{H}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , transuranic radionuclides  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$ , which are the main dose-related, and the most common radionuclides at the Semipalatinsk test site.

Work was carried out during the summer while stall maintenance of animals. Separate groups of animals (cattle, ovine, horses, and pigs) and birds were fed by the contaminated feed, soil, contaminated water, in some experiments, they were orally administered by the aqueous liquor with variety of radionuclides. Food, water and soil was prepared from the most contaminated sites that differ in technical character of radionuclide contamination. To control the intake of radionuclides in the body of animals and birds during the whole period of the experiment scientists recorded the number of eaten food, soil and water, as well as sampling of vegetation, soil, water, according to the scheme of the experiment

Terms of keeping animals and birds ranged from one to 120 days. At the end of each period, was made the slaughter of animals and birds by bleeding. On the analysis were collected: liver, kidneys, lungs, heart, spleen, muscle tissue, bone tissue, the fat tail, fur, skin, tongue, brain. In the future, selected and trained bioassays were transferred to spectrometric measurements.

As a result of this work there were obtained transition coefficients  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ ,  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$  in various organs and tissues of animals and birds. The parameters of radionuclide transfer in the "food-products", "soil-products", "water / liquor-products."

The obtained results confirm the existing data on the dynamics of accumulation and distribution of radionuclides  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the body of animals and birds. However, the resulting transition coefficients (R) were lower than the average values of R, represented in the IAEA database (TRS # 472, 2010). It is established that the bioavailability of radionuclides received with soil were much less than when entering the feed or water. Transuranic radionuclides incoming with soil, are absorbed less than when entering the feed. Under conditions of prolonged daily intakes of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  in the animal organism with the various components of the environment, the process of accumulation is observed in the liver, other organs and tissues of accumulation is not established. Tritium is distributed evenly in organs and tissues. It was found that for poultry products bioavailability  $^3\text{H}$ , received with food is more than  $^3\text{H}$ , received with air and water.

The obtained data on the distribution and accumulation of radionuclides in the organs and tissues of animals are the basis for the development of practical recommendations when deciding on the transfer of territories STS to economic use, as well as the development of a set of measures to reduce the radionuclide content in the products obtained in conditions of radioactive contamination.

In general, today, there were obtained adequately results for fat-tailed sheep the coarse wool breeds. The dynamics of radionuclide transfer  $^3\text{H}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$  in the organs and tissues of sheep and determine the nature of their distribution in the body, the transition coefficients received for these radionuclides in the "food-products", "soil-products."

Currently, there are made studies on parameters of radionuclide transfer in the pig and poultry production. These studies are expected to be completed next year. As a result of these studies, will be determined by the dynamics of radionuclide transfer and obtained coefficients of their transition to the organs and tissues of animals and birds in the system "water-products", "food-products", "soil-products."

Degree of scrutiny and completeness of research parameters of radionuclide transfer in the organs and tissues of large animals (cattle, horses) is less than studies with small animals. This is due, primarily, to great labor and financial costs. Despite this, studies and research in this direction are continued. Moreover, special attention is paid to transuranic radionuclides.

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# Study of aerosol $^{137}\text{Cs}$ activities around nuclear power plants in Slovakia

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Nuclear power plants (NPP) impact studies on the environment have been of many discussions during the last years, especially after the Fukushima accident. There are two NPPs under operation in Slovakia (central Europe), one in Jaslovské Bohunice and the second one in Mochovce. Both NPPs operate pressurized light water reactors of Russian origin (VVR-440) with 440 MW<sub>el</sub>. One of frequently studied radionuclides, which is produced in nuclear reactors and may be released to the environment is  $^{137}\text{Cs}$ . Most of the atmospheric  $^{137}\text{Cs}$  in this region comes, however, from the resuspension of the Chernobyl-derived  $^{137}\text{Cs}$ , as well as caesium produced during nuclear weapons testing, mainly during the 1960's (Kulan, 2006).

Atmospheric aerosol radioactivity has been continuously monitored at both NPPs. There is also a long tradition in monitoring of radioactivity in the Bratislava air by the Department of Nuclear Physics and Biophysics of the Comenius University. The first measurements of  $^{137}\text{Cs}$  on aerosols were carried out since 1976 (Povinec et al., 2012).

The objective of this work has been to study local trends and variations of anthropogenic  $^{137}\text{Cs}$  in the troposphere at each NPP location, to estimate a radiation impact of the NPPs on the local environment, and to compare the obtained results with the Bratislava monitoring station.

Atmospheric aerosols have been collected in weekly/monthly intervals on filters by pumping of large volumes of air (at least 50 000 m<sup>3</sup> per month). In most cases the used detection system, low-level gamma-ray spectrometers with HPGe detectors, reached only detection limits for weekly samples, therefore monthly intervals were chosen for the determination of  $^{137}\text{Cs}$  activities (Sýkora et al, 2008).

A typical trend in the  $^{137}\text{Cs}$  activity concentration in the air can be seen on Figure 1. In Bratislava aerosols, local maxima of concentrations are clearly distinguishable in winter seasons. Similar trends can be observed also in  $^{137}\text{Cs}$  concentrations at the Mochovce monitoring station. These maxima are connected with increased resuspension of particles from the soil due to enhanced windy conditions during the winter seasons.

Activity concentrations of  $^{137}\text{Cs}$  in Mochovce and Bratislava stations are approximately at the same level.

Such trend is not clearly visible in  $^{137}\text{Cs}$  concentrations measured at the Jaslovské Bohunice station, where the activities are higher in comparison to other locations. This increase of  $^{137}\text{Cs}$  in the atmosphere is connected with decommissioning of A-1 reactor after accident in 1977 (INES 4).

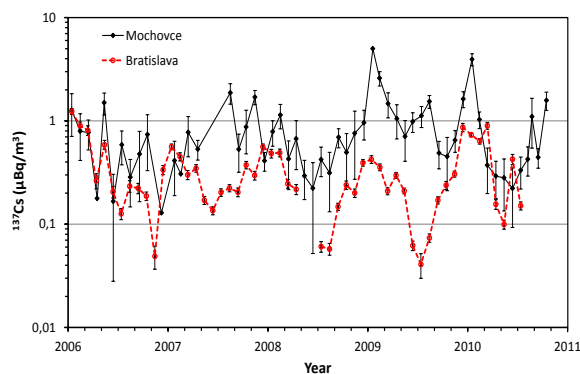


Figure 1. Comparison of monthly  $^{137}\text{Cs}$  concentrations in Bratislava and near the Mochovce NPP.

## Acknowledgments

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# IMPROVEMENT OF AN ATMOSPHERIC DISPERSION PROGRAM (SINAC)

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The SINAC (Simulator Software for Interactive Consequences of Nuclear Accidents) program system has been developed in the Hungarian Academy of Sciences Centre for Energy Research in the past decades and has been used to analyse the environmental consequences in case of an accident of a nuclear power plant.

Atmospheric dispersion, plume depletion by dry deposition and wash out, cloudshine and groundshine doses, dose consequences of inhalation and ingestion, early and late health effects are computed by the software. Effects of the introduction of countermeasures are also taken into account.

The SINAC program uses a Gaussian puff model for the calculation of atmospheric dispersion. In recent years three new modules have been developed regarding advection, dispersion and deposition for the new version of the SINAC program.

Instead of the previously used Pasquill categories the Monin-Obukhov length is applied to describe the stability of the atmosphere.

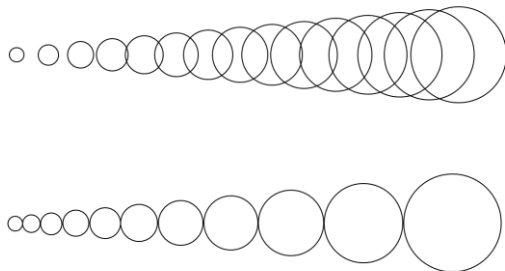


Figure 1. Comparison of the constant (above) and autoscaling (below) puff advection step options.

The SINAC program uses constant timesteps for propagating the puffs. In the new version, the autoscale method is applied, where the timesteps are proportional to the size of the puff. (Figure 1.) Thereby the accuracy of the mathematical calculations is independent of the wind speed and dispersion constant and at short distances they are better than those of the previous methods. The computational time is shorter at long distances keeping the same calculation accuracy.

In the previous version of SINAC a wind vector of only one vertical level was taken into account, in the new version dispersion is calculated based on an effective wind vector for all vertical levels in the planetary boundary layer throughout the mixing layer given by the numerical weather prediction model.

In the next phase the gamma-dose calculation modules will be revised.

Results of the new and the previous version of the SINAC were compared.

Detailed descriptions of the new models and the results of the comparison are reported.

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## Comprehensive analysis of 6 years of environmental dose data measured at the telemetric radiation monitoring stations around Paks NPP

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The Paks Nuclear Power Plant (NPP), located 5 km south of the city of Paks in Central Hungary, comprises four VVER-440/V213 type pressurized water reactor blocks. Continuous environmental dose rate monitoring is carried out with Bitt RS03/232 type proportional counters (Bitt detectors) and exposure of Al<sub>2</sub>O<sub>3</sub>:C thermoluminescent detectors (TLDs) of the PorTL portable TLD system is also performed in parallel at 9 type A radiation monitoring stations located at a distance between 1 to 2 km from Paks NPP. Bitt detectors and TLDs are installed at the same position at each type A station. The TLD at measurement point L25, located at a distance of 25 km, serves for providing background values. Further 11 gamma telemetric points, named as type G stations, are set up at the same distance from the NPP as type A points. For larger distances up to 30 km 14 type C TLD monitoring points are installed as well. Measurements with the two dosimeter systems have been performed regularly in parallel since 2009.

Both the Bitt detectors and the PorTL system are calibrated to provide environmental dose equivalent. In

order to compare data, first the monthly average dose rates were calculated. Analysing the measured data it has been found that the standard deviation for the Bitt detectors is  $\pm 7\%$ , and for TLDs it is  $\pm 9\%$  and  $\pm 11\%$  at type A and type C points, respectively.

Seasonal and geographical effects in the measurement data have been identified. A method based on double normalisation of data was devised for eliminating these effects from the raw measurement data. As a result, the detectable dose was reduced by a factor of 4 for the Bitt detectors and by a factor of 2 for the PorTL system.

The environmental monitoring system comprising Bitt detectors and the PorTL system proved to be stable and reliable over the years. A detailed analysis of the time series of data measured between 2009 and 2014, including a comparative assessment of dosimetry data is given in the present paper. The method for reducing the detectable dose and the lessons learnt are presented as well.

# In situ $\gamma$ -ray spectrometry in the deep oceans

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**Keywords:** *in situ method, marine radioactivity, MCNP5, NaI detector, deep ocean*  
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In-situ radioactivity measurements in marine environment are scarce, as most of the relative systems are not optimized and widely tested. The in-situ approach has been applied so far by installing the acquisition systems on large moored buoy networks [Tsabaris and Ballas, 2005; Tsabaris, 2008; Osvath et al., 2005]. The last years an underwater in-situ gamma-ray spectrometer has been developed to operate autonomously [Tsabaris et al., 2008] and to provide quantitative results [Bagatelas et al., 2010] for long term periods. The spectrometer provides activity concentration in absolute units (Bq/m<sup>3</sup> or Bq/l) for all gamma ray emitters in the energy range from threshold energy (50 keV) to 3000 keV.

The paper contains a study of the performance of sub-sea radioactivity configurations for monitoring radioactivity in the deep oceans. The design of detection system consists of a scintillation crystal of NaI(Tl) 3x3 inches and a stainless steel watertight enclosure. Other material are also studied for better efficiency in the high pressure. The materials are selected for the robustness and tolerance at the high pressures of the deep water masses. The design of the system provides maximum operational depth of 4500 m in order to cover all potential deployments at the Mediterranean Sea.

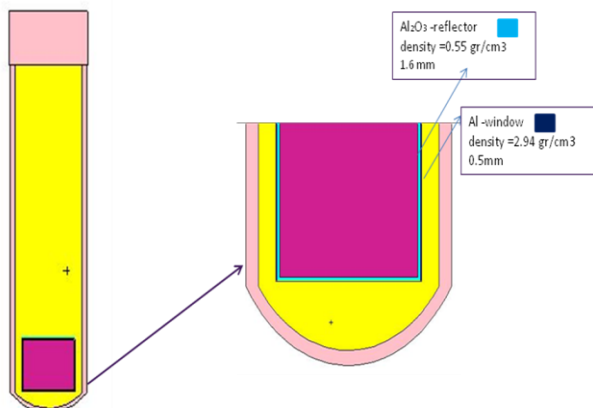


Figure 1. Simulation set-up

A theoretical study was performed by means of Monte Carlo (MC) simulation using the MCNP5 code. The first runs were dedicated to estimate the effective radius (the distance where a photon is fully absorbed) along with gamma-ray energy for two different thicknesses of the enclosure (0.7 and 0.9 cm) using as material stainless steel and 0.9 cm using as material of the enclosure Titanium. The whole simulation set up depicted in figure 1. The main difference between the proposed system

and the KATERINA system (Tsabaris et al., 2008), is the semi-spherical surface of the bottom of the system in order to reduce high pressures.

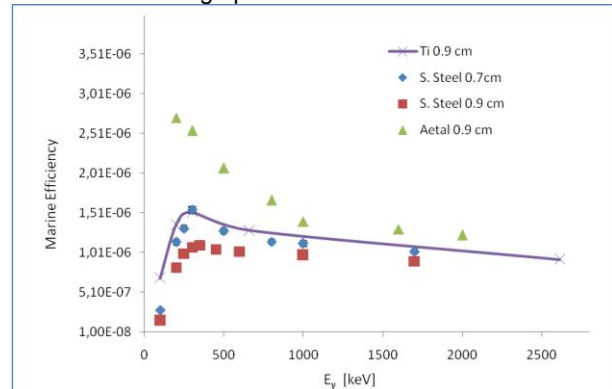


Figure 2. the efficiency along with gamma-ray energy for various enclosure materials and thickness.

The marine efficiency of the system was estimated for stainless steel and Ti using two different enclosure thicknesses running the MC simulations. More specifically, the values are represented for two enclosure thicknesses of stainless steel (0.9 and 0.7 cm) and 0.9 cm for Ti. The data are compared with previous data using acetal (0.9 cm) for pressures of intermediate water mass. The selection is performed taking into account the maximum operational depth and the minimum absorption of the gamma-rays into the material. The results exhibited similar values for energies above 1000 keV while the differences below this energy value are significant between acetal and the other materials. Titanium exhibits high performance at energies below 400 keV while above this energy the efficiency is similar between Ti (0.9 cm) and stainless steel (0.7 cm).

The sub-sea radioactivity detector has appropriate specification to be installed in any existing deep observatory for radon/thoron real-time monitoring. Furthermore, the in-situ detection system could be applied for detecting any anthropogenic radioactivity in deep marine compartments affected by nuclear activities (e.g. dumped nuclear wastes).

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# Environmental Radioactivity and Earthquake: a Possible Signature or the Real Trigger?

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Earthquake prediction is widely recognized as being among the most challenging scientific problems, both due to its societal relevance and to the intrinsic complexity of the problem, and it has been analyzed with several controversial discussions, debates, and reviews. Radon (Rn-222) as a possible candidate for earthquake's precursor has been studied for a long period, but there is no clear evidence that it is really a good precursor. It has been suggested as one of several possible early signals, and its groundwater anomalies associated with earthquakes and water-rock interactions were detected in several seismogenic areas worldwide indicating possible transport of radon through microfractures or the crustal gas fluxes along active faults. The physical processes associated with radon groundwater anomalies are based on changes of radon emanation rates occurring due to strain signal near the earthquake's nucleation point. Particularly, it is unclear its behaviour before, during and after the main shock, considering the consolidated scheme for radon release due to stress-strain processes in the rock. In the geological environment, the radon groundwater concentration depends on the isotopic abundance of its parent radionuclides (U-238 and Ra-226), and on their geochemical patterns with reference to environmental redox and pH characteristics. The geodynamic processes induced by earthquakes can modify radon migration patterns in groundwater as a potential indicator of strain. However, to predict the activity of radon in fractured lithologies is difficult and the measurement of radon concentration does not uniquely characterize the rock deformation or the chemical inhomogeneity, as well as its relationship with the transient crustal strain signals from 'aseismic' fault slip near the earthquake's nucleation point. Moreover, nontectonic factors related to variations of chemical and physical groundwater parameters may be of importance, requiring proper geological, hydrological and hydrogeological settings, so only the variations induced by stress-strain processes should be considered in evaluations.

In order to assess the utility of uranium isotopes as fluid phase earthquake precursors, uranium concentrations and U-234/U-238 activity ratios have been analysed as well as the mechanical behavior of uranium crystal metal, i.e. its structural behavior and its

phase stability, under several temperature and pressure has been investigated. Uranium enrichments in groundwater, that can be directly associated with the geodynamics of the earthquake, represent a much more precise strain-meter than Rn-222, whose presence could be modulated by uranium content during the preparation phase of the earthquake, and only successively released by microfracturing, during the main shock and aftershocks. Particularly, uranium groundwater anomalies can be used as a possible strain meter in domains where continental lithosphere is subducted. Furthermore, the structural behavior of uranium crystal under high temperature and high pressure conditions plays a key role in the earthquake's nucleation point due to uranium bulk modulus changes. Therefore, an answer to question "*uranium and earthquake: a possible signature or the real trigger?*" by numerical simulations as well as an improvement of the physical processes into the mechanics of seismic faulting have been introduced and discussed in our research activities on environmental radioactivity and earthquake.

## Acknowledgments

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# Determination of ppb levels of $^{238}\text{U}$ with low expanded uncertainties by pseudo-cyclic epithermal INAA using Compton suppression gamma-ray spectrometry

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Naturally occurring radioactive materials (NORM) have been present in our environment since the creation of the earth. The  $^{232}\text{Th}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  radionuclides and their decay products are of much interest because their emission of alpha- and/or beta-rays could be hazardous to living tissues.

Alpha- and gamma-ray spectrometry and inductively-coupled plasma-mass spectrometry (ICP-MS) are being used for the determination of NORM in diet and drinking water samples. For alpha spectrometry and ICP-MS chemical separations are needed to overcome interferences. Gamma-ray spectrometry on the other hand, as it has been employed in the past, is not sensitive enough for the low activity levels of NORM in food and drinking water samples; this lead to values reported in the literature as less than the detection limit. One of the objectives of this work was to develop a method by gamma-ray spectrometry for the determination of  $^{238}\text{U}$  and possibly  $^{232}\text{Th}$  in food items.

The first approach involved direct gamma-ray spectrometry and the reduction of the natural background radioactivity to improve detection limits using a Compton suppression system (CSS). To further improve these limits, a pseudo-cyclic epithermal instrumental neutron activation analysis (PC-EINAA) method in conjunction with CSS was developed for the determination of uranium and thorium.

Samples consisted of dried edible tissues of oyster and mussel. About 0.2 g of each sample was placed in 2-mL polyethylene vials and heat-sealed. Three different NIST SRMs, namely SRM 1566a (Oyster Tissue), SRM 1566b (Oyster Tissue), and SRM 1573 (Tomato Leaves) were used to validate the methods developed. Uranium and thorium comparator standards were prepared from ultrapure elemental standard solutions (SPEX industries Inc.). In order to obtain a similar sample geometry and density comparable to the real samples, the liquid solutions were spiked onto a high-purity sucrose base in a 2-mL polyethylene vial and dried under an infra-red lamp.

The samples, SRMs, uranium and thorium standards were irradiated at the DUSR facility in an epithermal neutron flux of  $2.5 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$  in the Cd-shielded site. The times for irradiation, decay and

counting for each cycle were 30 min, 3 min and 30 min, respectively. Four cycles were performed for each sample. Instead of using the same sample as commonly done in CINAA, four different sub-samples of each material were irradiated in this work to reduce undesirable background activities. Gamma-ray spectrum of each cycle was collected in a cumulative fashion.

The measurements were performed using a *p*-type HPGe coaxial detector (EG&G Ortec) in anticoincidence mode. This detector had a relative efficiency of 25% with respect to a standard NaI(Tl) detector, and a resolution (FWHM) of 2.0 keV at the 1332-keV photopeak of  $^{60}\text{Co}$ . The HPGe detector was surrounded by a guard detector consisting of a 10"x10" NaI(Tl) annulus with 5 photomultiplier tubes (PMT) and a 3"x3" NaI(Tl) plug with one PMT. The detector was connected to a digital gamma-ray spectrometer (ORTEC-DSPEC PLUS) and spectra were collected using Maestro-32 software.  $^{238}\text{U}$  and  $^{232}\text{Th}$  content was determined by the 74.6-keV (59.3%) gamma-ray of  $^{239}\text{U}$  ( $t_{1/2}=23.5 \text{ min}$ ) and the 86.5-keV (1.6%) of  $^{233}\text{Th}$  ( $t_{1/2}=22.3 \text{ min}$ ), respectively.

Uranium concentrations were measured in all samples and SRMs with good precision and accuracy. However, the thorium content could not be determined in either the SRMs or samples under the experimental conditions used. An  $L_D$  of 0.84  $\mu\text{g}$  and  $L_C$  of 0.40  $\mu\text{g}$  for thorium were calculated. It was concluded that a higher neutron flux is necessary for thorium determination by PC-EINAA-CSS in food samples. A radiochemical separation method was later developed for the determination of both uranium and thorium.

Every measurement is associated with some uncertainty. International Standards Organization (ISO) established general rules for the evaluation and expression of uncertainty in measurement in 1993 to give an indication of the quality of the results being reported. The expanded uncertainties associated with the determination of activities and concentrations of NORM by gamma-ray spectrometry were studied in this work. The relative expanded uncertainties at a coverage factor of 2 for uranium in reference materials was observed to vary from 2.3 to 4.2% for 1<sup>st</sup> through 4<sup>th</sup> cycle by PC-EINAA-CSS.

# Potassium to NaCl Ratios Assessments in the Dead Sea Ponds using In-Situ Gamma Spectrometry System

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**Keywords:** Natural Radiation, Gamma, Carnallite, NaI(Tl).

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The Dead Sea in Israel is an important source of minerals such as potash, a fertilizing chemical. In order to harvest the minerals, evaporation ponds system is maintained by streaming solution from the Dead Sea. Under a solution level in each pond, a wet Carnallite layer is embedded on the pond floor. Dry density and salt concentration determines the Carnallite quality, and is highly depended on the pond spatial and temporal conditions.

Carnallite molecule  $\text{KClMgCl}_2 \cdot 6(\text{H}_2\text{O})$  includes a potassium atom that naturally contains 0.11% abundance of the K-40 radioisotope, which emits 1463 keV gamma radiation. The Carnallite layers consist of a portion of dry Carnallite with a portion of NaCl with the solution phase. The purpose of this study is to establish an in-situ measuring system to probe inside the Carnallite layer in the ponds, and to determine the Potassium to NaCl concentrations ratios using the known gamma radiation emission of K-40.

An apparatus made of robust water proof encapsulated NaI(Tl) cylindrical detector (3 inch X 3 inch) was mounted. The system included a pulse height analyser with power supply (ScientiSpec) connected by USB cable to a laptop PC. The apparatus was set on a boat. The K-40 full photopeak net counts were collected within duration of 10 minutes for each position.

In order to predict the system feasibility we carried out Monte Carlo simulations. Monte Carlo simulations (MCNP) were preformed to assess the detector response to K-40 in homogeneous wet Carnallite medium. The spatial material spread was described in order to find the detection limits in terms of counts rate. We found that the detector response is maximal from certain Carnallite depth, due to its medium self-absorption (shown in Table 1).

Table 1. MCNP results of the detector response to K-40 gamma in a cylindrical Carnallite volumes (R = 30 cm).

Layer Depth (cm)	Relative counts	Cumulative counts	Response %
5	2.69E-03	2.69E-03	32.3
10	2.47E-03	5.16E-03	66.2
15	1.21E-03	6.36E-03	82.8
20	5.31E-04	6.90E-03	90.1
25	2.50E-04	7.15E-03	93.5
30	1.30E-04	7.28E-03	95.3

Presence of NaCl concentrations in the bulk, conducts to higher internal radiation attenuation. In order to extract the NaCl concentrations, several Monte Carlo simulations were carried out to extract the behaviour of the peak counts against excess of salt in a given Carnallite layer. The simulations results showed a 20% counts linear decrease per each increment of 26% NaCl (with  $R^2 = 0.9957$ ).

Finally, In-Situ radiation measurements were performed in several ponds, all with a 60 cm layer depth and a detector at 30 cm depth, each for 10 minutes period. In Figure 1 the results were plotted as net peak counts versus the NaCl concentrations. Clear linear dependence was obtained, therefore proving that the NaCl presence can be directly analyzed using peak counts in the range of 0 – 65% salt.

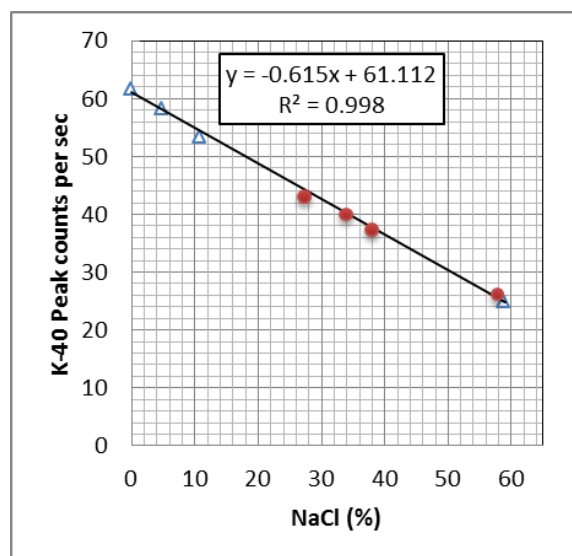


Figure 1. K-40 net peak counts versus NaCl concentrations in ponds' Carnallite layers.

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MCNP, Monte Carlo N-Particle Transport Code System, version 4C2, Technical Report LA-13 709-M, Los Alamos National Laboratory, NM, USA, 2001.

# In situ $\gamma$ -ray measurements on the seabed in two different marine environments

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**Keywords:** *in situ*  $\gamma$ -ray measurements, MCNP5, NaI detector, seabed, Black sea.  
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The interest for radioactivity monitoring in the marine environment continuously increases, mainly due to the presence of industries close to coastal zones. Laboratory-based activity measurements are widely performed on collected water and sediment samples, but are rather time consuming. A great benefit of implementing an *in situ* method instead, is the ability to obtain large scale data, using a cost-effective and easy to apply technique. An approach for *in situ* quantitative measurements on the seabed has already been developed Androulakaki *et al* (2015) by calibrating the NaI detection system KATERINA on full spectral range. Nevertheless, the determination of the detection efficiency remains a challenging problem, since it is site specific, related to the sediment characteristics.

In the present work, activity concentration measurements on the seabed were performed using the NaI underwater detection system KATERINA in two different marine environments, to study the influence of the detection efficiency with respect to the variations of the geological characteristics and to test the validity and limitations of the adopted approach. The first measurement was carried out at Lavrion area (Greece) and the second at Costinesti (Romania). In addition to the differences in the geological structure of the two regions, groundwater discharges (diffuse source) were also present in the latter.

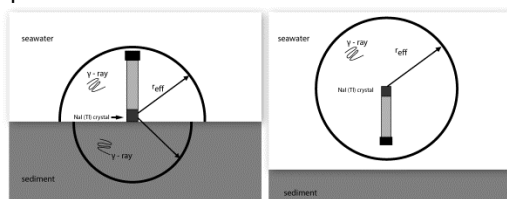


Figure1. *In situ* measurement a. on the seabed (left) b. on the seawater (right).

In the area of Lavrio, the detector was placed in close contact with the seabed (figure 1a) while in Costinesti the detector was placed a few centimetres above the seabed. Since the detector is surrounded by seawater, a second measurement in the seawater followed in both cases, with the detector being situated well above the seabed (figure 1b) in order to correct for events recorded in the spectrum originating from the seawater column. The detection efficiency was experimentally derived, in the energy range 350keV-2600keV, by calculating the count rate of the peaks from the acquired *in situ* spectra and using reference activity values determined with complementary laboratory based

$\gamma$ -spectroscopy measurements on collected sediment samples from the same site.

The Monte Carlo (MC) code MCNP5 was implemented to estimate the efficiency values for the measurements on the seabed in both study areas. All the crucial parameters that affect the detection efficiency, concerning the measurement set-up (distance between the seabed and the detector) and the physical characteristics (wet density, water content and composition) of the sediment were experimentally determined and implemented in the simulation. The key parameter, the effective sediment volume, defined as the volume from which  $\gamma$ -rays reach the detector, was calculated from the linear total attenuation coefficients of each  $\gamma$ -ray in the media (water, sediment). This is clearly illustrated in figure 2 for the 351keV  $\gamma$ -ray energy and for the three following conditions: the detector is placed in close contact with the seabed (figure 2a), the detector is placed 4cm above the seabed without altering the sediment characteristics (figure 2b), additionally the sediment wet density is increased by a factor of 1.5 to the value of 1.9 gr/cc (figure 2c). The initial effective volume gradually decreases while the shape changes from a hemisphere (figure 2a) to a steep sigmoidal (figure 2c), due to the significant change in the linear total attenuation coefficient of the two media (water, sediment).

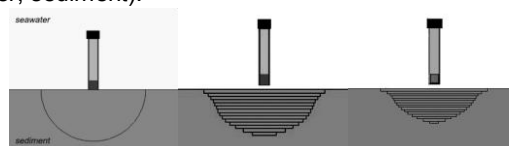


Figure2. Schematics of the effective volume for a. close contact (left) b. 4cm up and 1.4gr/cc c. 4cm up and 1.9gr/cc (right).

The MC results obtained implementing these complex geometries were found to be in satisfactory agreement with the experimentally derived efficiency values. This result triggered an extensive ongoing theoretical study, to systematically examine the influence of the detector efficiency to different sediment characteristics and measurement set-up. This theoretical study aims at limiting the necessity of the laboratory analysis for the quantification of the *in situ* measurements.

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## Advanced subsurface 3D electrical imaging and 4D time-lapse monitoring for groundwater contaminant remediation

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*Keywords: geophysics, subsurface contamination, monitoring, characterization.*  
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Electrical resistivity tomography (ERT) is a method of remotely imaging the bulk electrical conductivity of the subsurface. ERT has proven useful for groundwater applications because bulk conductivity is governed by many of the physical and geochemical properties of interest in groundwater contamination problems. Over the past 10 to 15 years, commercially available ERT data collection hardware has advanced rapidly, particularly for environmental applications, providing the opportunity to image the subsurface at high resolution over large spatial scales, and enabling subsurface processes to be monitored autonomously through time-lapse ERT imaging. However, the computational demands of processing such large data sets often prevent the full potential of ERT imaging provided by modern day ERT hardware from being realized.

To realize the full potential of ERT for large-scale subsurface characterization and time-lapse imaging applications, we have developed E4D, a distributed memory parallel inversion code designed specifically for

3D and 4D ERT imaging. We demonstrate several applications of high-performance ERT for imaging subsurface contamination or understanding properties and processes governing contaminant movement at the Hanford Nuclear Site. Past operations at Hanford released large amounts of highly saline low-level radioactive waste into the vadose zone through shallow infiltration galleries. There were also unplanned releases from failed single-shell, high-level radioactive waste storage tanks, although in much smaller quantities. The resultant highly conductive vadose zone contamination within Hanford's low-conductivity sands and gravels provides an excellent target for ERT imaging. Here we demonstrate 3D imaging with two massive ERT data sets collected to image deep vadose zone contamination that is beginning to reach the water table at the Hanford Site. Imaging results reveal previously unrealized detail concerning 3D contaminant migration and distribution, and are informing site operators about the origin of a nitrate groundwater plume emerging from beneath the B-Complex.

# Response study of a 1"x1" spectrometric dose rate LaBr<sub>3</sub> monitor for ground deposited artificial radionuclides

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**Keywords:** LaBr<sub>3</sub>, ambient equivalent dose, gamma spectrometry, variance reduction, Monte Carlo simulations  
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Spectrometric lanthanum bromide detectors (LaBr<sub>3</sub>(Ce)) are scintillation crystals with excellent energy resolution, i.e. at 662 keV ~ 3%. This makes them convenient for monitoring environmental radioactivity. Some countries such as Finland, are equipping their external monitoring stations close to NPP with this spectrometers (Salomaa and Sulonen (2011)). Furthermore, there are research projects going on in this area, such as the MetroERM European project (started June 2014) which aims, among others, to characterize novel gamma spectrometric detectors in order to analyze their response to ground contamination and radioactive clouds.

In order to carry out the characterization of these spectrometric monitors, one of the first steps and the main goal of this work is to study their response in case of ground contamination due to artificial radionuclides. The use of Monte Carlo (MC) codes has the advantages that it is possible to simulate any activity concentrations for different radionuclides and for several geometries of the site and the detector. However, one drawback of such methodology is the time consumption due to the geometry, i.e., an extensive source and a small detector size. Therefore, variance reduction techniques are strongly recommended. In this study, PENELOPE/penEasy (Salvat et al. (2011)) MC code with the reciprocal method as variance reduction technique (Zähringer and Sempau (1997)) is used. The main idea of the reciprocal transformation is to exchange the geometry of the source and the detector in order to work with a small source and an extensive detector. From MC simulation, the energy flux distribution that reaches the monitor is obtained, so the dose rate can also be derived. Then, this simulated energy flux distribution is convoluted using the energy response of the detector, which has previously been calculated with MC simulations, and the gamma spectrum from this detector is obtained. In order to validate the methodology a set of dose rate calculation have been compared with values obtained by Saito and Jacob (1995).

In the current study, this methodology has been applied to the station located at the roof of the building of South Campus of the Technical University of Catalonia (UPC) in Barcelona, with a soil radius of 10m and LaBr<sub>3</sub> detector at 1m height. Gamma spectra from <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>131</sup>I ground contamination, as they are common artificial radioisotopes in case of radiological nuclear accident, are generated with the above mentioned methodology. Then, these simulated spectra are added to background spectra measured at the station. Different activity concentrations for ground deposition are

evaluated and applied to different background scenarios, i.e., dry and rainy periods. The increase of background during rainy periods, due to <sup>222</sup>Rn progeny fallout, modifies the detection limit of the spectrometric monitors and should be analyzed.

Results show that spectrometric information could be - crucial for determining the presence of artificial contamination. As an example, figure 1 shows a waterfall plot with the results for 1kBq/m<sup>2</sup> ground deposition of <sup>134</sup>Cs for a 1-day period and including a rainy event. The ambient dose equivalent rate ( \* (10)) is also shown. Results with other radionuclides and activity concentration will be presented.

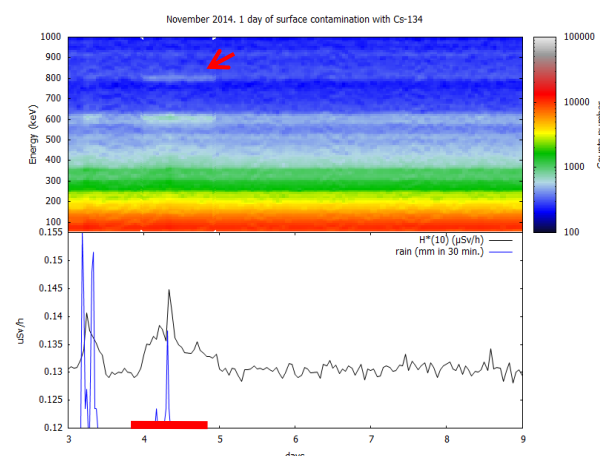


Figure 1 Waterfall plot shows the increase in counts due to ground deposition. Red arrow indicates the increase due to <sup>134</sup>Cs and red line the contaminated period.

## Acknowledgments

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# Long term monitoring of NPP Temelin (CR) outcome radionuclides using laboratory and in situ gamma spectrometry method

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Keywords: *biomonitoring, gamma spectrometry, Cs-137, NPP.*  
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Production of electricity in nuclear power plants represents a very small impact on the environment. The safe operation of each NPP is inspected using a number of measures and programs for monitoring a range of parameters inside NPP, and in its surroundings. Monitoring programs and their contents are accurately identified by the legislation, which is based on international recommendations and guidelines, prepared in part by IAEA, partly by the EU and state institutions.

The FNSPE (Faculty of Nuclear Sciences) became in 2000 (the year of the first NPP block activation), an independent observer of the environment in the vicinity of NPP Temelin, where the monitoring is carried out across a fixed point grid already in its sixteenth year.

The monitored area around NPP Temelin comprises 29 collection sites located along 8 profile radii reaching as far as 20 km from the Power Plant (the measurement points are located at a distance of 2, 5, 10, and 20 km from the Power Plant.) For monitoring, the following bioindicators were selected: forest humus, pine bark, *Pleurozium Schreberi* moss, edible mushrooms, and wild berries. Each two years the biomonitoring is supplemented by measurements determining the dosimetric and gamma-spectrometric characteristics of photon fields.

The laboratory gamma spectrometry measurements are conducted using coaxial HPGe detector (40%) in Marinelli geometry 0.6l. The time for dried samples measurement is 30,000 sec. on average. The program Genie 2000 is used for spectra processing and mass activities of presented radionuclides calculations.

The results of mass activities of <sup>137</sup>Cs (Bq/kg) in forest humus from 15 years monitoring time period enabled effective half-life calculation (see Figure 1).

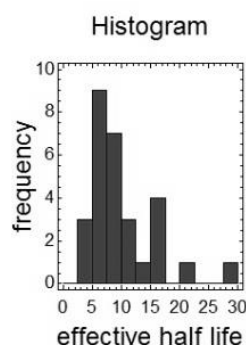


Figure 1. Effective half-life of <sup>137</sup>Cs frequency distribution based on 15 years of 29 locations monitoring

The presented wide set of measurements enables monitoring the behavior of radionuclide <sup>137</sup>Cs in bioindicators. In the year 2014-15 was determined the depth distribution of this radionuclide in forest humus and compared with results from the year 2001. An example is shown in Figure 2 and in Table 1.

Table 1. Example of the depth distribution versus effective half-life of mass activity <sup>137</sup>Cs (Bq/kg)

location	effective half life	depth of max <sup>137</sup> Cs
16	8.8 year	6-8 cm
17	22.4 year	2-4 cm
22	12.5 year	4-6 cm

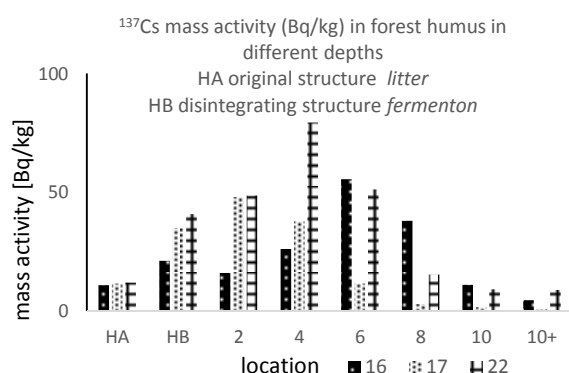


Figure 2. An example of the depth distribution of <sup>137</sup>Cs in forest humus 30 years after Chernobyl accident.

It is obvious that <sup>137</sup>Cs depth distribution differs at individual points, while the maximum concentration is in the range 2-8 cm. After 15 years of observing the value of specific activities <sup>137</sup>Cs it was noted, that these values are reaching the limit of detectability (apart from mushrooms and *fermenton* layer), using the laboratory gamma-spectrometric methods available.

## Acknowledgments

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# In-situ $\gamma$ -ray measurements for background environmental studies and calculation of external doses rates in Brazil

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**Keywords:** In-situ  $\gamma$ -ray measurements, environmental measurements, natural radiation, external dose rates.

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The determination of ambient dose rate which the population is exposed is the main objective of any environmental monitoring programs. The sources of radiation which contribute to this rate are of natural origin (cosmic radiation, radionuclides of the U and Th series, Radon and Thoron and  $^{40}\text{K}$ ) and artificial (fall-out of nuclear explosions, Chernobyl and Fukushima accidents, release of radioisotopes in nuclear facilities, use of radioactive sources in industries and in nuclear medicine).

The final objective of this work is start acquire enough data to compose a radiation background map of Brazilian territory. The first survey areas are the known Brazilian anomalies and areas that are used for nuclear and radiological facilities. This map will be used to estimate public exposure and to have a database for impact assessment from environmental releases of radionuclides and also for accident emergencies, in support for decision-making processes.

The measurements are performed using an **Spectrometric Portable Radiation Scanner (SPRS)** AT6101C Scanner, from ATOMTEX® and a **Spectral Advanced Radiological Computer System (SPARCS)** from U.S Department of Energy – International Emergency Management and Cooperation. These multichannel NaI(Tl) systems can be used in vehicles, boats, aircraft and helicopter and the AT6101C can also be used in "man borne" configuration (the main use).

Average dose rates for Fortaleza is  $80 \pm 23$  nSv/h, for Vitoria is  $96 \pm 33$  nSv/h and for Angra dos Reis is  $147 \pm 16$  nSv/h. These results are then compared to previous results on other coastal urban towns (Rio de Janeiro, Niterói and Salvador, Martin Island in Itaguaí Bay near the future Brazilian Navy Base for Nuclear Submarines), and with the high background coastal area of Guarapari town.

Table 1. Values found for Ambient Dose Rate (nSv/h).

Town	Average	Min	Max	Reference
Fortaleza	80	19	190	[1]
Sauípe	47	19	104	[1]
Salvador	43	39	52	[2]
Vitória	96	54	257	[1]
Niterói	105	75	151	[2]
Rio	96	75	126	[2]
Angra	147	121	172	[1]
Sorocaba	75	30	100	[4]
Iperó	43	31	63	[4]
Martin Is.	40	19	364	[3]
Amazon R.	10	10	20	[3]

Two other areas of primary interest are the site of Brazilian Multipurpose Reactor (RMB) in the city of Iperó and a long survey over rivers of amazon rainforest.



Figure 1. Detail of survey over Itaguaí Bay and Martin Island. The high values inside the island are due decorative stones in fences and houses. Units in nSv/h.

Although it is not appropriate use a general media for dose rates without consider the different pavements and use of the land we can assume that the values measured can represent very well the radiation field. The presence of NORM, mountains, and big structures of concrete increase the dose.

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# The analyses of radiation results from various geographical settings by employing a self-developed in situ gamma ray detection system

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**Keywords:** *in situ gamma detection, naturally occurring radionuclides, phosphate mine, beach sediment.*  
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Gamma ray spectroscopy as a survey tool has been successfully applied in the fields of morphology, geology and mineral exploration. Gamma ray surveys are regularly done in hugely varying geographical environments. A robust and portable gamma ray detection system, the GISPI (Gamma In Situ Portable Instrument), was developed and applied for this purpose. This system acquires gamma ray spectra, extract radionuclide concentrations and finally interpolate data to provide radionuclide concentrations and produce maps while on location. Two recently completed radionuclide mapping projects will be discussed.

The GISPI was employed to map nuclide concentrations in coastal sediment along various beaches in and around Saldanha Bay, located on the West Coast of South Africa. A mathematical model was developed and implemented in order to analyze the in situ gamma ray spectra. Several other techniques were applied to obtain GIS overlays that could be used to investigate various geophysical features. The results demonstrated direct relationships between radionuclide concentrations, grain size of beach sediment and the intensity of wave action (see Figure 1).

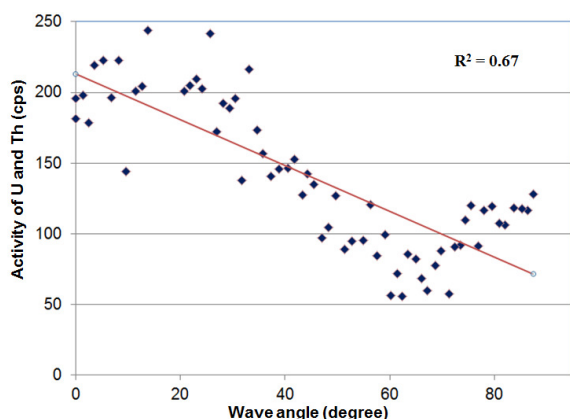


Figure 1. A graph of the combined uranium and thorium counts as function of wave action on a beach.

Conclusions on coastal and beach formation processes, based on the results of the distribution of various radionuclides were also made. A model was finally developed to describe coastal sedimentation and beach erosion patterns that occur due to anthropogenic interference.

High levels of uranium and its radioactive progeny like radium is normally associated with phosphate mining. The GISPI was therefore employed to determine

the concentrations of naturally occurring radionuclides at a spend phosphate mine. The concentrations of radium, thorium and potassium were measured and plotted. High concentrations were confined to specific areas of the mine. The effective dose for the various areas of the mine was also estimated (see Figure 2). Final deductions were made as to the origins and impact of the radiation.



Figure 2. A Google Earth image with an overlay indicating the effective dose rate at the phosphate mine.

The different geographical settings demanded various means of transport which included motor vehicles, quad motorcycles and transporting the system on foot. The system however performed well in these challenging environments and various other projects are consequently planned. Conclusions will finally be made on the success of methods and equipment that was utilized in the studies. The results indicated several environmental concerns in the areas that were investigated and these will also be highlighted.

## Acknowledgments

This work was supported by the National Research Fund of South Africa.

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## Radioecology for the 21<sup>st</sup> century: trends for the future

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Born together with the development of the civil use of nuclear energy, radioecology has evolved as a multidisciplinary scientific discipline aimed at describing, understanding and predicting the fate of artificial and natural radioactivity in environmental systems, its impact on man and the environment in order to support human and ecological risk assessment, and biogeochemical processes by means of tracer studies.

During the recent decades, radioecology priorities have been shaped by the Chernobyl accident with large emphasis dedicated to unravel transfer of radionuclides within the environment towards man in order to support human radiation protection issues. More recently, under the notable incentive of the International Union of Radioecology (IUR), priorities have been shifted to address protection of the environment *per se* in addition, as a response to the parallel development of concerns about sustainability development and global environmental issues.

Obviously, the most recent Fukushima accident is now influencing the directions most needed for radioecology in the 21<sup>st</sup> century. IUR, the largest worldwide network of radioecology specialists, has identified three major challenges to be tackled.

The first one relates to the need to develop worldwide networking of efforts, with clarification of the respective roles of currently up to 15 regional/topical networks contributing to the synergy between science and expertise in radioecology. The IUR FORUM launched last year has collectively agreed to tackle this challenge. This is important because accidental radioactive contaminations are susceptible to affect large areas across several countries where administrative borders are no longer meaningful, and also because scientific controversies are still on-going, especially about potential ecological impact, therefore undermining public trust in nuclear generation as a CO<sub>2</sub> free energy source.

The second one consists in promoting a move towards a more ecocentric vision for radioecology, such as to allow for true ecological risk assessment that would address directly the actual targets of environmental protection, populations and ecosystems. Currently, the biocentric approach based upon "reference animals and plants" (as per ICRP) cannot resolve the whole issue without addition of a further "ecosystem approach".

Finally, one should not ignore the practical challenges faced at Fukushima featuring especially: the need to revisit decontamination and remediation techniques in order to solve acute waste-related issues, the need to develop better capacity for assessing impacts on the marine environment, and finally, as already mentioned above, building better scientific consensus on low dose effects in chronic exposure to radiation on animals, plants and their ecosystems.

# Eight years of $^{210}\text{Pb}$ concentrations monitoring at two sites in the Mediterranean region: seasonality and identification of potential sources

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Keywords:  $^{210}\text{Pb}$ , seasonal variability, back-trajectories, PSCF.

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Due to its production mechanism and environmental fate,  $^{210}\text{Pb}$  radionuclide has been proven as a useful tracer of continental air masses. Moreover,  $^{210}\text{Pb}$  is also, together with its decay product  $^{210}\text{Po}$ , the natural radionuclide with the highest contribution to the annual effective dose received by the population for ingestion and inhalation of uranium and thorium series radionuclides other than radon (UNSCEAR, 2000). Due to its potential impact on the dose rate, it is necessary to identify and investigate the sources and meteorological conditions promoting  $^{210}\text{Pb}$  increases.

With this aim, this work 1) characterizes and compares the seasonal and interannual variability of  $^{210}\text{Pb}$  concentrations, and 2) identifies the geographical location of sources of high  $^{210}\text{Pb}$  concentrations at two monitoring sites located in the densely populated Mediterranean region, El Arenosillo (37.10 N; 6.70 W; 40 m a.s.l.; Spain) and Mt Cimone (44.20 N; 10.70 E; 2165 m a.s.l.; Italy). Besides the long-term contemporary monitoring of  $^{210}\text{Pb}$  concentrations (2004-2011 period), by means of  $\gamma$ -spectrometry carried on  $\text{PM}_{10}$  filters, the two sites were chosen to represent different sampling areas in the Mediterranean basin: in fact, while El Arenosillo is a coastal low-elevation station considered as a background station (Lozano *et al.*, 2012), Mt. Cimone is a high-altitude WMO-GAW station representing free-troposphere background conditions during most of the year (besides summer months when there is an influence of the innermost layer due to higher mixing height and higher convection) (Tositti *et al.*, 2014).

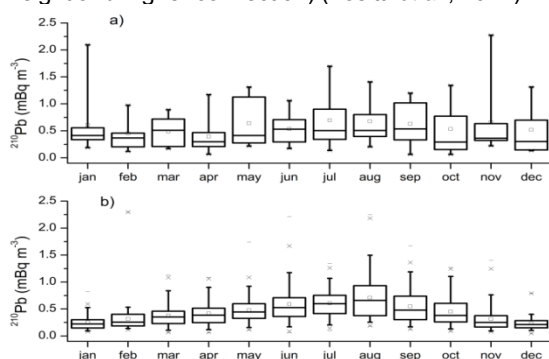


Figure 1 (a,b). Boxplots showing the seasonal variability in the 2004-2011 period at the two sites: a) Huelva; b) Mt. Cimone.

Figure 1(a,b) reports boxplots comparing the seasonal variability of  $^{210}\text{Pb}$  concentrations at the two

sites. It is clear that while at Mt. Cimone a clear and distinct seasonal pattern is present, at El Arenosillo the seasonal behaviour is less clear, showing that the variability at Mt. Cimone is governed by synoptic and regional conditions, as opposed to El Arenosillo where the  $^{210}\text{Pb}$  concentrations are governed by a combination of synoptic, regional and even local conditions. Similar to other mid-latitude sites, at both sites the maximum  $^{210}\text{Pb}$  concentration appears during the warm season, even though with a smoother pattern in El Arenosillo, as compared to Mt. Cimone where a definite peak in August appears. During the cold season, the difference between the two sites is well marked, due to the different altitude of the two sites.

A statistical method based on back-trajectories calculated during the sampling period, namely the Potential Source Contribution Function (PSCF) method of Ashbaugh *et al.* (1985), was applied to gain information of the geographical location of sources affecting  $^{210}\text{Pb}$  increases at the two sites (Figure 2).

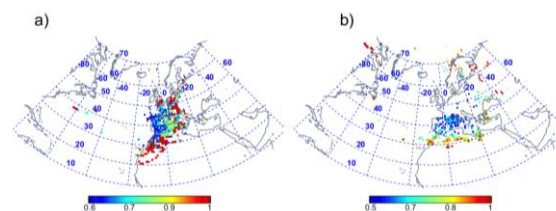


Figure 2 (a,b). Conditional probability maps of  $^{210}\text{Pb}$  sources at Huelva (a), and Mt. Cimone (b), choosing as high concentrations  $^{210}\text{Pb}$  values equal to or above the 50<sup>th</sup> percentile.

Clear differences are shown. At El Arenosillo, the regions contributing mostly to high  $^{210}\text{Pb}$  concentrations are clearly northern Africa (Sahara desert) and central Europe, while at Mt. Cimone the sources of  $^{210}\text{Pb}$  appear as very dispersed and very difficult to determine, being nevertheless northern Africa the most important contributor. In both cases, the Mediterranean sea appear to behave as an important reservoir layer affecting  $^{210}\text{Pb}$  concentrations.

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# Measurement of environmental radioactivity using a cumulative gamma radiation dosimeter; Fertilization-induced changes in young fruit trees

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Keywords: environmental radiation, radiocesium, monitoring, dosimeter, fruit tree  
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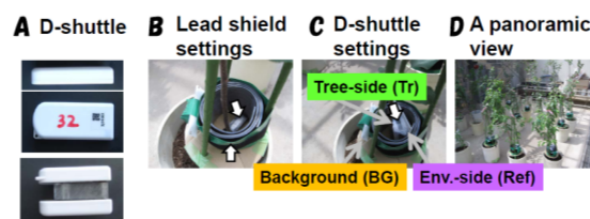
The accident at the Fukushima Dai-ichi Nuclear Power Plant following the huge earthquake on March 11, 2011, dispersed radionuclides across large areas of northern Japan (Morino et al., 2011). Since then, many decontamination programs have been implemented hurriedly in both residential and agricultural areas, including crop fields and orchards. However, these programs cannot negate concern regarding the potential contamination of produce (MOE, 2012; Forestry Agency, 2012). In particular, the long-term growth to harvest required by fruit trees increases the concern regarding the potential for secondary contamination from soils via the root system or inner circulation. Thus, a continuous program of monitoring is indispensable. However, the majority of current monitoring systems are based on invasive techniques using a Ge-semiconductor detector. Although such systems provide accurate data, they require physical samples for every measurement, disturbing the targeted trees and/or the environment. In addition, implementing such systems is labour intensive and expensive and thus, there is a need for cheaper and more efficient alternative methods.

Here, we present our concept for a simple method of on-site monitoring together with the results of a trial measurement. The method is based on the measurement of cumulative gamma radiation doses using a recently commercialized personal dosimeter (D-shuttle; Chiyoda Technol. Corp., Japan). D-shuttle can measure doses at very fine resolution ( $>0.01 \mu\text{Sv h}^{-1}$ ) and store the data for more than 12 months. It is noteworthy that the data are recorded as hourly cumulative doses and can be read at discretionary durations.

If D-shuttle could separately detect a dose from a targeted object against the background doses (BG), it would be applicable for monitoring the long-term transition of radionuclides in the environment. In the present trial, the targets were young trees of peach (*Prunus persica* cv. Akatsuki), apple (*Malus pumila* cv. Fuji), and persimmon (*Diospyros kaki* cv. Hachiya). These trees were grown over several months in pots containing soils contaminated by radiocesium and both with and without potassium fertilizer. D-shuttles were set to the tree trunks both with and without a lead shield to separate the BG and targeted doses (Fig.1).

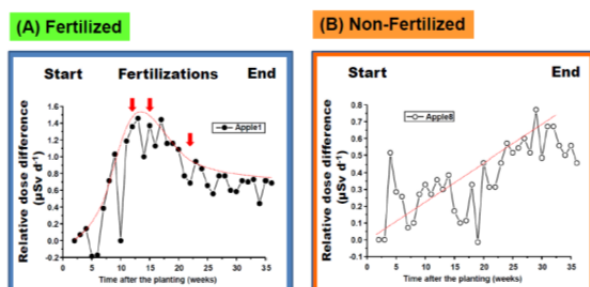
The results demonstrated that D-shuttle clearly detected the targeted doses from the trees and reflected specific changes in the doses induced by fertilization and/or seasonal effects. Even though further investigation (e.g., specific shielding effects of trees) is required to establish the specific correlation between the

cumulative dose and radioactivity concentration, D-shuttle could be considered a useful tool for monitoring long-term changes of radionuclide distributions in the natural environment (Fig.2).



**Fig. 1 D-shuttle (A) and the settings (B-D)**

A, The outside view of D-shuttle (L68 mm, D 32mm, W 14mm, 23 g); B, Two lead shields (base, 15 mm thick; and circular, 9-12 mm thick) were set at the bottom of trunk. C, One and two D-shuttles each was set at the outside and inside of the circular shield, respectively. Another lead shield (15 mm thick) separated the inside D-shuttles.



**Fig. 2 A typical change in the apple plantlets**

After increase, the dose of the fertilized plantlet gently reduced, whereas that of the non-fertilized plantlet increased.

Figure 1. Theoretical and experimental values of  $^{137}\text{Cs}$ .

## Acknowledgments

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# Signature of a Sudden Stratospheric Warming in the near-ground $^7\text{Be}$ flux.

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**Keywords:** Vertical atmospheric coupling, Sudden Stratospheric Warming, cosmogenic isotope.

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We present here a study of the impact of one Sudden Stratospheric Warming (SSW) upon the atmospheric vertical dynamics based on  $^7\text{Be}$  measurements in near ground air, using both numerical and conceptual. In late September 2002, an unprecedented SSW event occurred in the southern hemisphere (SH), causing changes in the tropospheric circulation, ozone depletion and weakening of the polar jet in the mesosphere. There is an observational evidence suggesting that anomalies in the stratosphere play an important role in driving tropospheric weather producing tropospheric changes that can persist for up to 60 days in NH and up to about 90 days in the SH, as observed after the 2002 SSW (Thompson et al., 2005).

Radioactive environmental techniques for tracing large-scale air-mass transport have been applied in studies of atmospheric dynamics for decades and they are becoming more and more precise due to the improvement of the instrumental sensitivity and associated modeling. Temporal variations of the cosmogenic  $^7\text{Be}$  (half-life time  $T_{1/2} = 53.22 \pm 0.06$  days) concentration in the near-surface atmosphere can provide information on the air mass dynamics, precipitation patterns, stratosphere-troposphere coupling and cosmic ray variations.

The present study is based on an analysis of  $^7\text{Be}$  concentration measured in near-ground air in the city of Angra dos Reis, Rio de Janeiro state, Brazil (23°00'S 44°19'W, geomagnetic cutoff rigidity  $P_c = 11.6$  GV) between 1987 and 2009. The mean  $^7\text{Be}$  activity concentration during the entire period was  $0.84 \pm 0.44$  mBq.m<sup>-3</sup> and there was a clear outlier ( $\sim 3.5$  mBq.m<sup>-3</sup>) in 2002-2003. A spectral analysis shows an absence of a significant solar cycle modulation imprints over this Angra's  $^7\text{Be}$  data (Leppänen et al. 2010), which is due to the high local geomagnetic cutoff rigidity ( $\sim 12$  GV) and the complex regional synoptic-scale climatic systems. Pacini et al. (2011) also found that the depositional flux and the air-mass transport are the dominant sources of  $^7\text{Be}$  variability in the low atmosphere in RJ area. Moreover, using a simplified tropospheric  $^7\text{Be}$  model deposition based on a two-layer transport model, Pacini (2011) reported that the occurrence of strong downward air flux leave an imprint of the 3D motion of air masses to the near-ground air  $^7\text{Be}$  data.

In this work, we have further developed the two-layer model described in Pacini et al. (2011) by adding one more layer: the lower stratosphere (LS). In normal

conditions, the contribution of the low-stratospheric  $^7\text{Be}$  to the near-ground isotopic variability would be very small, since the mean residence time of the  $^7\text{Be}$  in the stratosphere is about two years. On the other hand, stratospheric source can be crucial for the SSW event. The  $^7\text{Be}$  concentrations for the near-ground air computed by the three-layer model for both scenarios (equilibrium and after SSW) are plotted in Figure 1, along with the  $^7\text{Be}$  data measured in Angra between Jun 2000 and Jun 2004.

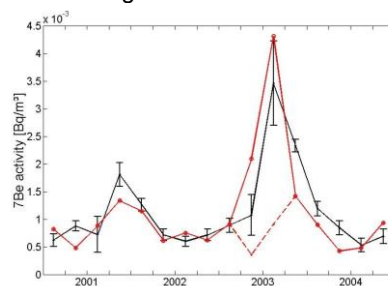


Figure 1 – Measured  $^7\text{Be}$  activity concentration data for Angra (black) along with the results obtained with the three-layer model (red) in the equilibrium conditions (dotted) and considering the stratospheric intrusion in 2002.75 (continuous line).

This result indicates that a strong stratospheric air intrusion happened after the SSW and induced a downward flux of stratospheric aerosols from the LS to the ground level lasting several months after the SSW peak, showing that its tropospheric consequences can be much larger than it is usually considered.

## Acknowledgments

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# SPECIATION OF TRITIUM IN SOIL AT THE TERRITORY OF THE SEMIPALATINSK TEST SITE

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**Keywords:** tritium, speciation, soil, the Semipalatinsk Test Site.  
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Nuclear explosions at the Semipalatinsk Test Site (STS) have resulted in radioactive contamination of environmental objects. One of the most widely spread artificial radionuclides at the STS territory is tritium. Concentration of tritium both in water and soil achieves values of several hundreds of thousands Bq/kg (Aidarkhanov et al., 2013). Herewith speciation of this radionuclide in soil is hardly studied. It is known that tritium in water can be contained as free water, in organically bound and tightly bound form (Lopez-Galindo et al., 2008). Speciation of tritium in soil is the main parameter, characterizing its migration.

This work is aimed to determine the speciation of tritium in soil at different areas of the STS, differing each from other in relief, soil-climatic and ecological conditions.

For research purposes samples of soil were collected at different radiation-dangerous objects of the STS (crater of the «Atomic» lake, external water reservoir, Shagan river). Research methodology based on consecutive extraction of each form of tritium from soil sample: surface adsorbed water, interlayer water, crystallized water, organically bound tritium and tightly bound tritium. Different speciation of tritium, contained in form of free water was determined by distillation at different temperatures. Bound forms of tritium were determined using the method of autoclave decomposition with variations of physical and chemical conditions. As a result of experimental works concentrations of tritium in obtained samples have been determined using the method of liquid scintillation spectrometry.

As a result it was found that there is some difference in speciation of tritium in soil of

radiation-dangerous objects of the STS. In samples of soil in the area of Shagan river tritium mainly contains in form of surface adsorbed water (93-99%). In the northeastern part of the «Atomic» lake prevails tritium in organically bound form (81-85%) while in the opposite direction in the south-eastern part prevails tritium in form of surface adsorbed water (38-41%) and tightly bound form (34-45%). In soil samples collected around the perimeter of water reservoir organically bound 3H is the major of tritium occurrence form (55-94%).

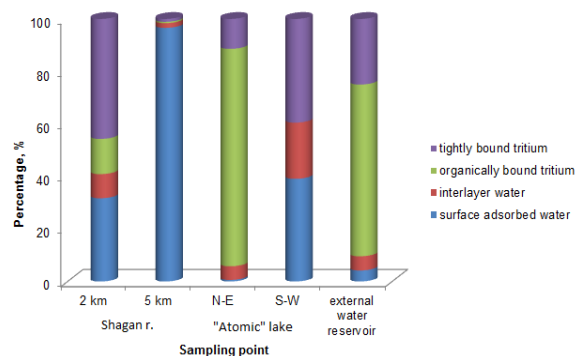


Figure 1. The distribution speciation of tritium

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# Background reduction in a gamma spectrometer setup with an active shielding

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The construction of an active shielding for a commonly used detector system is described and tested. Changes in detection limit and measurement time are presented and discussed.

In our laboratory we use several HPGe Detectors for gamma spectroscopy. For this study a characterized extended Range coaxial germanium detector the Canberra model GX5019 was chosen. The gamma spectrometer is protected by passive shielding (100 mm of Pb, 1mm of Cd, 10 mm of Cu and 5 mm of Plastic). This setup is mainly used to measure food products for the German IMIS network and sediment dating with <sup>210</sup>Pb. The samples often contain very small amounts of radioactive isotopes and low detection limits are required.

Moving the lab underground would be a good option to decrease cosmic background, unfortunately that is not possible at our university. As an alternative an active shielding was constructed.

Active shielding systems have shown good results in the past as shown by Hurtado *et al* (2006), Pointurier *et al* (1996) even for laboratories above ground. This study shows that active shielding systems can easily and at moderate cost be integrated into day to day measurements to improve detection limits or reduce measurement times.

The active shielding consists of two 50x100x10 cm<sup>3</sup> plastic scintillation detectors mounted on top of the detectors lead castle. Summed up signals from the scintillators are processed with timing signals from the HPGe into coincidence signals. The coincidence signals are used as anti coincidence gates.

A major part of the background counted in a gamma detector is created by cosmic radiation, mainly through muon interactions with the lead castle and consequent neutron induced reactions. On sea level approximately 75% (Núñez-Lagos *et al*, 1996) of the cosmic radiation are muons. With a cos<sup>2</sup> angular distribution for the zenith angle  $\theta$  of the incoming muons on sea level (Particle Data Group, 2014), approximately 80% of the incoming muons pass through our top mounted scintillators, before reaching the detector. Depending on the combined efficiency of the setup they can be detected and recognized as coincidences.

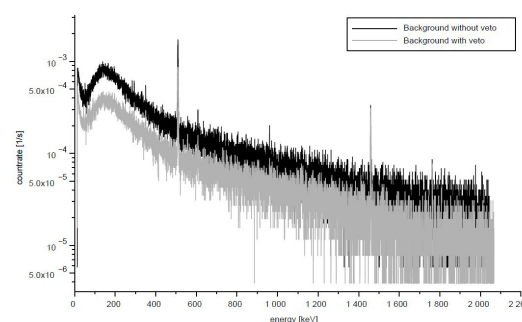


Figure 1. Long time background count rates with and without active shielding

Tests with applied active shielding show promising results of approximately 50% reduction for total background count rates. This improves the MDA for <sup>210</sup>Pb and <sup>137</sup>Cs in our setup by 25% and 28% respectively.

	cps	MDA <sup>210</sup> Pb 46.5keV [Bq/kg]	MDA <sup>137</sup> Cs 661.7keV [Bq/kg]
No veto	1.30	0.855	0.025
Veto	0.66	0.642	0.018

Table 1. Total Count rates (30keV-2MeV) and minimum detectable activity

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# Detection of Fukushima-released radioisotopes in Málaga (South Spain)

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Keywords: Fukushima, <sup>137</sup>Cs, <sup>134</sup>Cs, <sup>131</sup>I, gamma spectrometry.

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This work presents measurements of the effect of the atmospheric radioactive release from the Fukushima Dai-ichi nuclear power station at Málaga, on the southeastern coast of Spain (36° 43' 40" N; 4° 28' 8" W). The sampling site is one of the environmental radioactivity monitoring network stations operated by the Spanish Nuclear Security Council (CSN), under a cooperative agreement with the University of Málaga through the Environmental Radioactivity Research Group.

Activity concentrations of several man-made radionuclides (such as <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs) were detected along the Iberian Peninsula from March 28<sup>th</sup> to April 7<sup>th</sup> 2011 (Baeza et al., 2012; Piñero and Ferro, 2012). The first detection of Fukushima-released radioisotopes in Spain were found in the high-volume filter collected in Seville during the period 14<sup>th</sup>-21<sup>st</sup> March, in Cáceres during the period 15<sup>th</sup>-2<sup>nd</sup> March, while in Barcelona and Málaga, radioisotopes were found during the period 23<sup>rd</sup>-30<sup>th</sup> March.

The analysis of back-trajectories of air masses allowed us to support that the levels of man-made radionuclide activity concentrations in the south of the Iberian Peninsula came from the accident produced in the nuclear power plant of Fukushima. The pathway followed by the radioactive plume from Fukushima into Málaga was deduced through back-trajectories analysis, and this fact was also verified by the activity concentrations measured of those radionuclides reported in other places crossed by this radioactive cloud. This qualitative analysis was done using the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) developed by the NOAA Air Resources Laboratory (ARL). Calculations were done for three altitudes above ground level, at 500 m, 1500 m, and 3000 m, to acquire information on air masses circulating at lower and upper levels.

Our sampling point is located on the flat roof of the SCAI building at the University of Málaga, at a height of 10 m above the ground and approximately at 5 km from the coastline, near the airport and surrounded by roads with traffic exhaust. Aerosols, atmospheric bulk deposition and food samples were analyzed to determine the radioactive activity. Airborne particulate samples were collected on G3 polypropylene square filters (440mm side) using a high-volume sampler (ASS-500 station) with an air-flow rate, in normal conditions, of approximately 700 m<sup>3</sup> h<sup>-1</sup>. Deposition samples were collected in an area of 1 m<sup>2</sup> using a collector that is a slightly tilted stainless steel tray and filling 60 L polyethylene vessels with bulk deposition. Mixed diet

samples were collected on different days. Measurements by gamma-spectrometry were performed to determine the activities of the samples using an intrinsic REGe detector.

The highest concentrations of <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs in air samples were 670±17.8 µBq/m<sup>3</sup>; 91.0±1.78 µBq/m<sup>3</sup>; 76.2±1.11 µBq/m<sup>3</sup>, respectively. After precipitation on March 23<sup>rd</sup>-31<sup>st</sup>, the maximum concentrations of <sup>134</sup>Cs and <sup>137</sup>Cs were detected in bulk deposition samples, 28.7±4.8 mBq/L; 36.2±9.4 mBq/L respectively, also in wet deposition samples in the same period, maximum concentration of <sup>131</sup>I 67.4±4.0 mBq/L were detected. As a consequence, <sup>137</sup>Cs was transferred to the human food chain: 32.5±19.9 Bq/kg person.

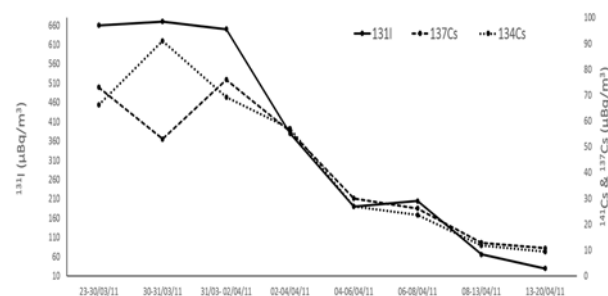


Figure 1. Temporal evolution of <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs levels in aerosols collected at Málaga station.

The traces of <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs detected in the different samples were very low after the Fukushima nuclear accident. Consequently, the associated fallout had negligible radiological consequences in our sampling area.

## Acknowledgments

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# Design of the negative ion cooler at MALT

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Isobaric separation is quite important to improve the measurement accuracy and background for accelerator mass spectrometry (AMS). In order to suppress isobaric interference, several methods have been proposed and utilized. Actually, a gas counter or a gas-filled magnet has been conventionally used (Matsuzaki, 2008; Aze, 2007). In these devices, nuclides of interest can be separated from isobars by difference of interaction between ions and gas materials.

In the 1980's, Berkovits et al. tried to remove stable isobars before acceleration with laser light (Berkovits, 1989). In this technique, difference of the electron affinity (EA) of nuclides of interest and interference materials is used. Assume the EA of the nuclides of interest (EA1) is higher than the EA of the isobaric nuclides (EA2). When laser light, which energy is higher than EA2 and smaller than EA1, is injected into the negative ion beam containing both of a target and isobaric nuclide, only negative isobar can be selectively neutralized by photo detachment reaction. In a usual AMS system, ion beam is injected into the accelerator with the bending magnet. Therefore, if ion beam is interacted with laser before injecting the accelerator, neutralized isobars cannot enter the accelerator. Consequently, only nuclide of interest can be injected into the accelerator and isobaric suppression can be effectively achieved. However, due to the limited laser performance, the laser-ion interaction time was too short to suppress isobar sufficiently at that time. Therefore, this technique has not been in practical use yet.

Recently, as laser improved in quality and the way to increase the laser-ion interaction time effectively was proposed, development of isobaric suppression system is going on (Liu, 2012). The apparatus for laser-ion interaction is called an ion cooler and consists of an electrode for deceleration, a quadrupole electrode to trap ions, and an electrode for re-acceleration after ions pass

through the quadrupole. The quadrupole region is filled with a buffer gas and ions collide with gas molecules, which results in the deceleration of ions and the long interaction time. This photo detachment system can remove isobaric interference in AMS measurements for nuclides, such as Cl-36 (EA=3.62eV) with S-36 (EA=2.08eV), Ni-59 (EA=1.156eV) with Co-59 (EA=0.661eV) (Liu, 2012). Furthermore, even if the EA of the nuclide of interest is lower than the EA of the isobaric nuclide, photo detachment would be useful by converting the nuclides into the molecular ions and reversing these electron affinities.

This new isobaric suppression system will be introduced into Micro Analysis Laboratory, Tandem accelerator, University of Tokyo (MALT) in the future. As a preliminary step of the introduction, we designed the ion cooler including electrodes for deceleration and re-acceleration and a quadrupole electrode and calculated ion behavior in the ion cooler with SIMION (Dahl, 2000) (Fig.1). In this study, the calculation results and the development of the negative ion cooler at MALT will be presented.

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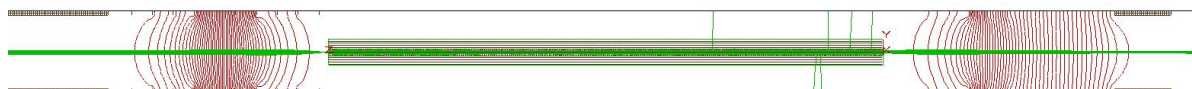


Fig. 1 Ion behavior in the ion cooler



# Analysis of the relationship between climatic phenomena and atmospheric variability of $^7\text{Be}$ and $^{210}\text{Pb}$ concentrations in northeastern Spain

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**Keywords:**  $^7\text{Be}$ ,  $^{210}\text{Pb}$ , Teleconnections index

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The inter annual and inter seasonal variability of the long period time-series of atmospheric concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$ , measured at a coastal and middle latitude site, has been analyzed by wavelet transform and coherence methods to evaluate their relations to climate indexes such as the North Atlantic Oscillation (NAO) (Rodó, Baert and Comín, 1997) and the Western Mediterranean Oscillation (WeMO) (Martín-Vide and Lopez-Bustins, 2006).

The radionuclides concentrations were measured by  $\gamma$  spectrometry analysis of filters collected weekly pumping a large volume of air at the Institut de Tècniques Energètiques of the Universitat Politècnica de Catalunya (INTE-UPC) in Barcelona (lat. 41.39N; long. 2.18E; 15 masl) since 2001 (Valles et al., 2009). In Figure 1 the  $^{210}\text{Pb}$  and  $^7\text{Be}$  long time series measured from 2001 and 2014 is presented with their monthly rolling means.

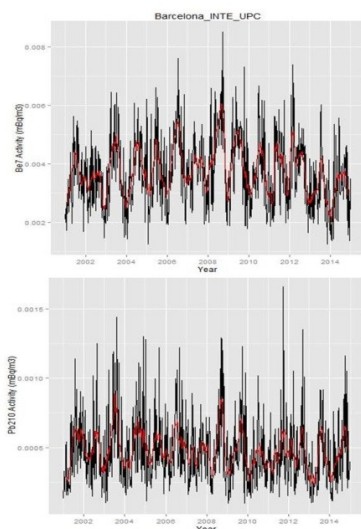


Figure 1. Weekly Time series of  $^{210}\text{Pb}$  and  $^7\text{Be}$  measured in Barcelona and their rolling average.

The monthly NAO index data used is based on the difference of normalized sea level pressure (SLP) between Lisbon/Portugal and Stykkisholmur/Reykjavik. Positive values of the NAO index are associated with stronger-than-average westerlies over the middle latitudes, more intense weather systems over the North Atlantic and wetter/milder weather over Western Europe (Hurrell et al., 2009). The Western Mediterranean Oscillation (WeMO), is only defined within the synoptic

framework of the western Mediterranean basin and its vicinities. In Figure 2 the trend level plots of monthly NAO and WeMo index variability is presented.

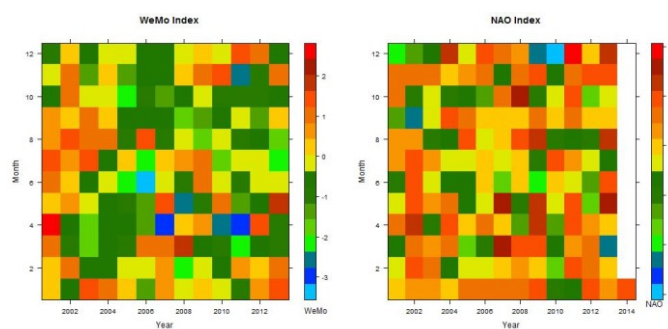


Figure 2. Level plot of the Wemo and NAO during period 2001-2014

Early data analysis show highest concentrations of  $^7\text{Be}$  observed during spring and summer seasons and mainly due to tropospheric masses intrusion when Wemo index is mainly neutral.  $^7\text{Be}$  strongly decreases during winter when Wemo index uses to be negative. First Wavelet coherence method results show a modulation of  $^7\text{Be}$  and  $^{210}\text{Pb}$  by WeMo while it is less affected by NAO variability. Nevertheless, a spatial analysis including more Spanish  $^7\text{Be}$  and  $^{210}\text{Pb}$  stations could be useful to understand these climatic correlations over Spain.

## Acknowledgments

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# Surface activity levels of $^7\text{Be}$ , $^{210}\text{Pb}$ , $^{40}\text{K}$ and other atmospheric species and the influence of African air mass intrusions

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Keywords: Atmospheric radionuclides; Aerosols; Back-trajectories; Dust intrusions  
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Natural radionuclides from terrestrial and upper atmospheric sources and of anthropogenic origin are widely used as tracers to examine atmospheric processes relevant to air quality and climate and to validate atmospheric models.  $^7\text{Be}$  and  $^{210}\text{Pb}$  are elements which may be used as indicators for air mass flow and vertical interchange dynamics between the stratosphere and troposphere.  $^{40}\text{K}$  is found in most types of soil and can easily be transported by re-suspended material. Concentrations of  $^7\text{Be}$ ,  $^{210}\text{Pb}$ , and  $^{40}\text{K}$  in air are continuously monitored using a high-volume air sampler and a high resolution gamma-ray spectrometer in Málaga, a Mediterranean coastal city in South Spain located to receive pollution from different sources by means of air mass transport (Gordo *et al.*, 2015). The high frequency of mineral dust plumes blown across from Africa to the Iberian Peninsula significantly affects dust levels in the atmosphere (Rodríguez *et al.*, 2001). Therefore, it is of great interest to analyse and quantify the African dust loading over the region due to the impact it has on air quality.

This work presents results from a 2-year sampling campaign carried out to provide information on the frequency and impact of these African dust episodes on the concentrations of radioactive matter and non radioactive matter in the lower atmosphere at this Mediterranean coastal site. The sampling campaign was started on January 2013 and continued until December 2014. Aerosol samples for radiometric measurements were collected at the University of Málaga (36° 43' 40" N; 4° 28' 8" W) over 3-day periods (occasionally 4 days) using a high-volume sampler fitted with square polypropylene filters (440x440 mm<sup>2</sup>). In addition to the normal operation, we added samples collected according to dust forecasts provided by CALIMA network ([www.calima.es](http://www.calima.es)). All the radiometric measurements were performed by low-level gamma spectrometry with a coaxial-type germanium detector and it was calibrated using certified reference gamma ray cocktail. Additionally, concentrations of particulate matter fraction PM10, SO<sub>2</sub>, NO<sub>2</sub>, CO and O<sub>3</sub> were obtained from the monitoring station "Carranque" (36° 43' 40" N; 4° 28' 4" W), belonging to the Atmospheric Pollution Monitoring network managed by the Environmental Health Service of the Andalusian Government. The identification of African events was confirmed by means of 4-day backward trajectories (at 500, 1500, 3000 m.a.g.l.) computed using the HYSPLIT model (Draxler and Rolph, 2013) and BSC-Dream8b dust images.

Basic statistics on the overall radionuclide dataset are summarized in Table 1. Higher average concentration values were found for the three

radionuclides in the set of samples collected under the influence of African dust events.

Table 1. Statistical summary of the activity concentrations of the three gamma radionuclides.

Radionuclides (Bq/m <sup>3</sup> )	AM	GM	Max	Min	SD
$^7\text{Be}$ ( $\cdot 10^{-3}$ )	2.89	2.71	6.18	0.50	0.98
$^{210}\text{Pb}$ ( $\cdot 10^{-4}$ )	0.57	0.47	1.58	0.08	0.35
$^{40}\text{K}$ ( $\cdot 10^{-5}$ )	1.77	1.52	6.84	0.29	1.12

The correlation patterns of the three radionuclides with meteorological variables and with the concentrations of the other atmospheric species were examined, both on the total set as well as on dusty and non-dusty basis. Significant correlations were found for  $^7\text{Be}$ ,  $^{210}\text{Pb}$ , PM10 and temperature during non-dusty days. Under the influence of African dust outbreaks, correlations among the different variables changed, being  $^7\text{Be}$  positively correlated with  $^{210}\text{Pb}$ , PM10, temperature and O<sub>3</sub>. Additionally, significant correlations were found for  $^{40}\text{K}$  with PM10, SO<sub>2</sub>, NO<sub>2</sub> and O<sub>3</sub>, during days with dust episodes. This study highlights the importance of multidisciplinary monitoring campaigns and the need to include radiotracer measurements in atmospheric studies. The use of natural radiotracers and other atmospheric species provides a solid background for the characterization of atmospheric transport as well as in supporting source apportionment of pollutants.

## Acknowledgments

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# Study of iodine dynamics in soil based on the analysis and modelling for the I-129 depth profiles before and after the FDNPP accident

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**Keywords:** soil, iodine dynamics, iodine-129, Accelerator Mass Spectrometry, Fukushima Daiichi Nuclear Power Plant  
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Iodine is one of halogen elements universally existing on the earth environment (Muramatsu, 1998). It is biophilic element and often concentrated near the large carbon reservoir like methane hydrate or natural gas. Recent studies have revealed it also take a important role for the climate (Kupper, 2008). However the details of the global cycle of iodine has not yet fully understood. This is partly because iodine takes many chemical forms in the environment different from other halogen elements. Also because it is strongly affected by the bioactivity.

According to Muramatsu, 1998, nearly 70% of total iodine exists in the oceanic sediments. One of the important source of the oceanic sedimentary iodine is vast amount of bioactivity of marine biology. But iodine component transported by the terrigenous sediment should not be small. On the land environment, iodine is concentrated in the soil nevertheless the bedrock as a mother material of soil typically contains fairly small amount of iodine. Most of iodine in soil should have obviously been transported during the soil formation by more or less bioactivity. However the kinetic property of iodine is still very poor.

To address this problem, a long-lived radio-nuclide <sup>129</sup>I (half life = 1.57E+7 y) is very useful. Iodine-129 is naturally produced in two ways. One is by the interaction between the cosmic ray and Xe in the atmosphere. The other is as a spontaneous fission of <sup>238</sup>U in the earth crust. However the amount of these natural production is not much, consequently the isotopic ratio of <sup>129</sup>I/<sup>127</sup>I is estimated to be less than 1E-12. On the other hand, a vast amount of <sup>129</sup>I has been produced artificially after 1950's. Firstly it is by the atmospheric nuclear bomb tests and followed by the operation of spent fuel re-processing plants. These anthropogenic <sup>129</sup>I has been released into the environment and the isotopic ratio (<sup>129</sup>I/<sup>127</sup>I) had raised up in several orders. Far from currently active fuel reprocessing plants like Asian region, the anthropogenic <sup>129</sup>I is mainly transported from the atmosphere. Especially on the land, <sup>129</sup>I falls on the ground by dry and/or wet deposition. I then diffuse deep into the soil. The depth profile of <sup>129</sup>I reflects the on going diffusion. We can derive the iodine kinematic information in the soil from the depth profile. Fukushima Daiichi Nuclear Power Plant (FDNPP) released high dense radioactive nuclides regionally including <sup>129</sup>I which provides sharp isotopic spike into the environment.

Soil core samples (30cm long) were collected from Ibaraki and Saitama, middle region in Japan, before the FDNPP accident. After the FDNPP accident, soil cores were collected from the region affected the accident in Fukushima. Each soil core was cut at depths with interval of 1.5 to 3.0 cm. Each cut piece was dried and homogenized. Iodine in the soil was extracted by the pyrohydrolysis method according to Muramatsu, 2008. Iodine concentration was measured by ICP-MS. I-129 concentration was determined by AMS (Accelerator Mass Spectrometry) at MALT, The University of Tokyo for the extract iodine with carrier iodine. From these two analytical results, the isotopic ratio <sup>129</sup>I/<sup>127</sup>I was calculated.

From the analytical experiments, depth profiles of <sup>129</sup>I for the soils before and after the FDNPP accident were obtained. Figure 1 shows the depth profiles of <sup>129</sup>I in the soil of before (not affected by) and after (affected by) the FDNPP accident. From the detailed analysis and modeling, iodine dynamics information will be obtained.

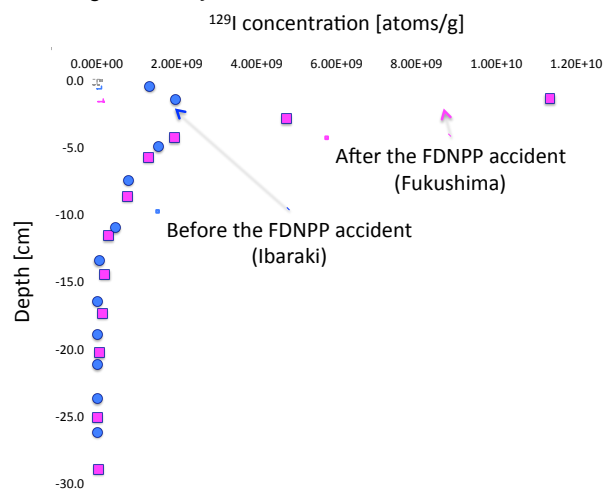


Figure 1. Iodine 129 depth profiles before and after the FDNPP accident

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# Concentrations of radiocesium for seafood in Korea, 2011-2014

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Keywords: Radiocesium, Seafood, Korea, Fukushima accident.  
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The public concern on the safety of seafood contaminated with radionuclides has dramatically increased in Republic of Korea since the March 2011 Fukushima accident. The activity concentrations of radiocesium were monitored to assess the level of artificial radionuclides contamination in major seafood organisms caught in Korean coastal waters as well as the imported ones from other countries during 2011 ~ 2014.

Target samples were total 33 kinds of seafood organisms including 2 species of macroalgae, 2 species of crustaceans, 5 species of mollusks, 2 species of cephalopods, 1 species of surface water fish, 16 species of mesopelagic fishes and 5 species of bottom fishes. Samples were purchased from domestic fishery market and import agents with the information of date and sea area collected.

Sample not less than 10 kg was prepared for each species. Individual of sample were divided into skin, muscle and internal organ in laboratory. Combined each part was freeze dried and grinded. A portion of homogenized sample was carbonized in electric furnace at lower than 450°C. The activities of gamma emitting radionuclides were counted using gamma counter (Canberra HPGe Well-type detector). A part of muscle for most species and whole body for macroalgae, mollusks and surface water fish (anchovy) were used for counting gamma emitting radionuclides. The correction for the efficiency of detector and geometry was done using several certified reference materials of IAEA/RGU-1, RGTh-1, RGK-1 and IAEA-414.

The concentrations of  $^{137}\text{Cs}$  in samples caught in Korean coastal waters were in the range of 0.03 ~ 0.26 Bg/kg-ww and  $^{134}\text{Cs}$  in these samples was not detected. These values are similar with the concentrations of  $^{137}\text{Cs}$  in marine organisms monitored before 2011 Fukushima accident (Choi *et al.*, 2008). The concentrations of  $^{137}\text{Cs}$  detected were 0.06 ~ 0.07 Bg/kg-ww in macroalgae, 0.05 ~ 0.18 Bg/kg-ww in crustaceans, 0.04 ~ 0.10 Bg/kg-ww in molluscs, 0.4 ~ 0.14 Bg/kg-ww in cephalopods,  $0.09 \pm 0.08$  Bg/kg-ww in surface water fish, 0.06 ~ 0.26 Bg/kg-ww in mesopelagic fish and 0.11 ~ 0.25 Bg/kg-ww in bottom fish, respectively. The concentrations of  $^{137}\text{Cs}$  in 6 species of marine organisms imported from other countries were in the range of 0.02 ~ 0.28 Bg/kg-ww except mackerel ( $2.68 \pm 0.27$  Bg/kg-ww) which was collected in the Pacific Ocean of Japanese eastern coast in June 2012.  $^{134}\text{Cs}$  in this mackerel was also detected as much as  $1.94 \pm 0.44$  Bg/kg-ww.

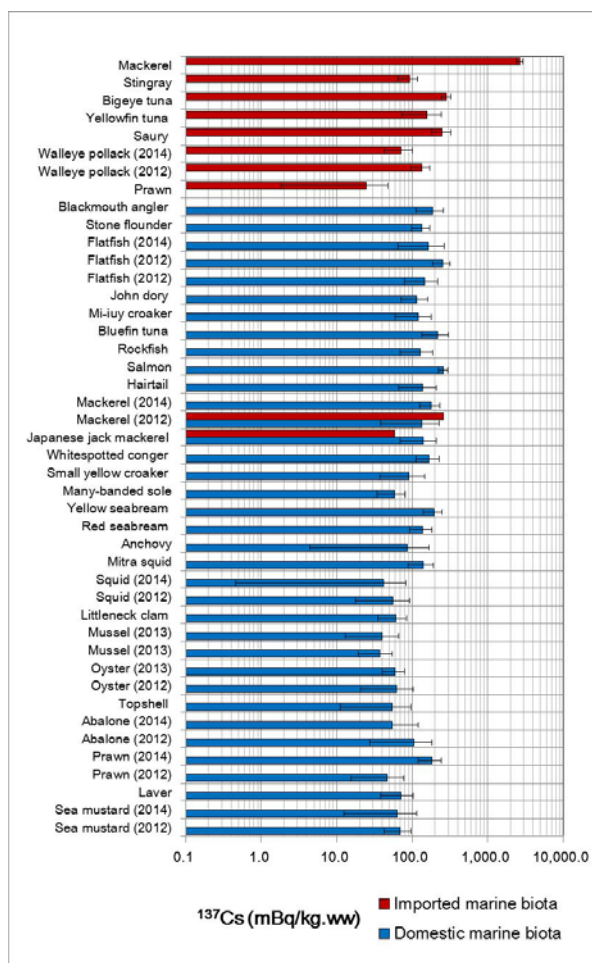


Figure 1. Concentrations of  $^{137}\text{Cs}$  in seafood which Korean consumed mainly, 2011-2014.

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## Acknowledgments

This work was supported by Korea Institute of Ocean Science Technology Grants (PG48740 and PE99232).



## INFLUENCE OF RADIONUCLIDES $^3\text{H}$ ON MORPHO-ANATOMY INDEX OF PLANTS *ACHNATHERUM SPLENDENS* AND *ELYMUS ANGUSTUS* ON THE STS

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Keywords: STS,  $^3\text{H}$ , tritium, plants, morpho-anatomy index.

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One of the problems of the Semipalatinsk Test Site (STS) is the contamination of  $^3\text{H}$  of natural areas. This area is ecosystem of the river Shagan. The concentration of  $^3\text{H}$  in surface waters is  $4 \times 10^5$  Bq/kg. This value exceeds the permissible level for drinking water (10 Bq/kg) 50 times. Radionuclides  $^3\text{H}$  is interesting because it is capable of high speed included into biological processes. So that radionuclides may be a powerful mutagenic factor.

Studies carried out on a section of high- $^3\text{H}$  in the river. The number of research sites was 10 pieces. The distance between the sites ranged from 40 to 1000 m. At each areas selected by two plant species: *Achnatherum splendens* и *Elymus angustus*. The test samples shown overground parts of plants. Determination of tritium activity in free water performed using a liquid scintillation spectrometer TRI-CARB 2900 TR. The detection limit was 10 Bq/kg, an error was less than 20%. Morphological studies were included: measuring plant height, leaf length and reproductive organs (ear or panicle). Anatomical measurements were included: parameters stems (stem diameter, the thickness of the epidermis and sclerenchyma, the area of vascular bundles located in the parenchyma) and leaves (the thickness of the adaxial (upper) and abaxial (lower) epidermis, mesophyll and the area of vascular bundles). Anatomical time preparations prepared by a sledge microtome with a freezing device, sections consisted in fixing balm. Total trained more than 200 temporary micropreparations. These preparations measured under the microscope Micros 300 V21 with increasing 4, 10, 20, 40 and 100x.

In result, the range of specific activity  $^3\text{H}$  in plants ranged of 1 kBq/kg to 100 kBq/kg. With an increase in the specific activity of  $^3\text{H}$  from 1.5 kBq/kg to 95 kBq/kg observed decrease in the diameter of the stem *Achnatherum splendens* from 3500 m to 1900 m (Figure 1). In some cases, the same content of  $^3\text{H}$  in plants are observing as the maximum and minimum values for the different tissues. Thicknesses of sclerenchyma stems from *Elymus angustus* with specific activity  $^3\text{H}$  100 kBq/kg are in the ranges of 120-160 m and 200-230 m. In *Achnatherum splendens* thickness of sclerenchyma stems with specific activity  $^3\text{H}$  35 kBq/kg is in the range 180-260 m and 350-500 m. In *Elymus angustus* at maximum specific activity  $^3\text{H}$  100 kBq/kg values stem diameter in the range of 1600-1700 m and 2500-2650 m. Changes in the tissues that form the leaf blade in both species are within the error limits.

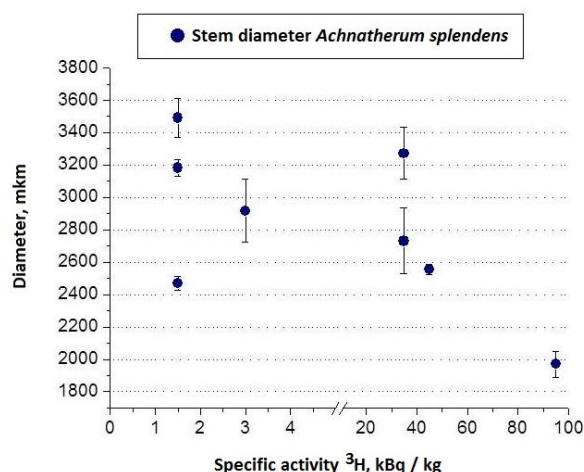


Figure 1. Changes in the diameter of the stem depending on changes in the specific activity of  $^3\text{H}$  in free water plants.

As a result of impact radionuclide  $^3\text{H}$  on plants, changes are observed values of individual stem tissues *Achnatherum splendens* and *Elymus angustus* on the anatomical level. Changes in the leaf blade are minor and are within the error limits for both species. Changes in plants in the investigated concentration range  $^3\text{H}$  not detected on the morphological level.

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# Benchmarking of Monte Carlo simulations for a Lanthanum Bromide ( $\text{LaBr}_3$ ) detector

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Keywords:  $\text{LaBr}_3$  detectors, gamma spectrometry, MCNP simulations  
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In order to improve Environmental Radiation Monitoring Networks, new systems with spectrometric capacity have been studied, including Lanthanum Bromide detectors ( $\text{LaBr}_3$ ).

Lanthanum Bromide ( $\text{LaBr}_3$ ) detectors are scintillators that offer significantly better resolution (<3 percent at 662 keV) than sodium iodide ( $\text{NaI(Tl)}$ ) detectors.

The first part of the study dealt with the acquisition of spectra in our laboratory, figure 1, using a 1.5 x 1.5 inches  $\text{LaBr}_3$  detector and a  $^{137}\text{Cs}$  point source.



Figure 1: Experimental arrangement

In parallel, using the Monte Carlo code MCNP-5 the same situation was simulated: the detector geometry, the point source and the constructive details of the laboratory, such as the floor, the walls, detector and source holders. Gaussian energy broadening is used to model the resolution of the  $\text{LaBr}_3$  detector.

The results for the first spectra (experimental and simulated) comparison were that they were quite similar.

However, the effects on spectra of those materials present in the lab (walls, floor, table, holders and so on) were very important, and so, the table where the detector and source are placed as well as the holders have been replaced with smaller devices made of lighter material (polymethyl methacrylate (PMMA) in order to minimize scattering.

After the installation of the new elements a calibration was made using  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  point sources.

The distance between the detector window and the source was 35 cm and the experimental spectra of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  had at least 10000 counts at the central channel of the peaks.

At the same time, the new elements had been implemented for the MCNP simulation.

The measurements and the simulated results for  $^{137}\text{Cs}$  are shown in figure 2.

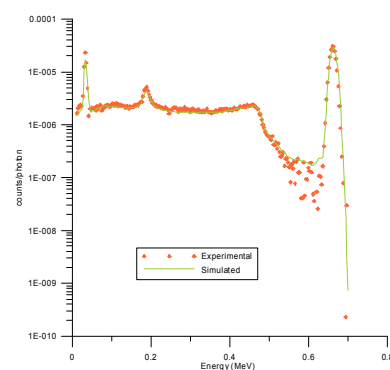


Figure 2: Experimental & simulated spectrum of  $^{137}\text{Cs}$ .

We can conclude that measurements and simulated results have an excellent agreement in the three cases.

As a next step different incidence angles at the detector will be considered, in order to study their influence on spectra.

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# Time lag between the tropopause height and the levels of $^7\text{Be}$ concentrations in surface air

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Keywords:  $^7\text{Be}$ , atmosphere, tropopause, Solar cycle, correlation coefficient

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The aim of the present work is to define the time delay (time lag) between the changes of the tropopause height and the  $^7\text{Be}$  concentrations in surface air at Northern latitudes over  $60^\circ\text{N}$ .

According to our knowledge, in mid latitudes there is a strong positive correlation between the seasonal changes of the tropopause height and the concentrations of  $^7\text{Be}$  in surface air and in case of  $40^\circ\text{N}$  has defined a time-lag between tropopause height and  $^7\text{Be}$  surface concentrations of 3-4 days.

In this work we examine the influence of tropopause height on  $^7\text{Be}$  concentrations in surface air at latitudes above  $60^\circ\text{N}$ . The concentration of  $^7\text{Be}$  at near surface air has been determined over the year 2009, at three different locations in Finland: Ivalo ( $68^\circ 64'\text{N}$ ,  $27^\circ 57'\text{E}$ ), Rovaniemi ( $66^\circ 51'\text{N}$ ,  $25^\circ 68'\text{E}$ ) and Kotka ( $60^\circ 48'\text{N}$ ,  $26^\circ 92'\text{E}$ ). Year 2009 was a year of solar minimum, ie. a year of high production rate, and at the same a period when the cosmogenic flux was stable. So it was the ideal period to study atmospheric changes and reveal the differences in  $^7\text{Be}$  fluctuations due to any meteorological and seasonal variations.

For our analysis, the tropopause height was determined for a small shell that covers each one region for year 2009 daily.

The equation used in order to find the height of the tropopause is given here.

$$\text{TropoHeight} = \frac{Z_1 + Z_2}{2}$$
$$Z_1 = \frac{287 \text{Temp}_{li}}{9.81 \log\left(\frac{\text{Pr}_{li}}{\text{Tropopres}_i}\right) + \text{GH}_{li}}$$

The number 1 refers to the one isobaric surface and (i) runs from 1 to 365 days of 2009. The same is for the second surface. (Temp) and (Tropopres) refer to the temperature and pressure of the air at the tropopause levels. (GH) and (Pr) refer to the geopotential height and pressure of the closest isobaric levels. Holding the same column of data for  $^7\text{Be}$  we calculate the correlation coefficient (R) for each new column of daily data of the tropopause height. The new columns are created by going back in time with a step of one day in order to find how many days we have to wait until the concentrations of  $^7\text{Be}$  responds to the elevation of the tropopause height.

Analysis gave that at latitudes over  $60^\circ\text{N}$  the correlation between the tropopause height and  $^7\text{Be}$  concentrations is weak.

For Ivalo and Rovaniemi it was found that changes in the daily surface concentrations of  $^7\text{Be}$  lag the changes in the elevation of the tropopause by four days (Fig. 1).

In Kotka station, the influence of tropopause height on the surface concentrations of  $^7\text{Be}$  is the weakest (Fig. 1). In Kotka region it seems that the influence of air masses from the East has greater influence on  $^7\text{Be}$  concentrations instead of the influence of the tropopause height.

It seems that in high studied latitudes, the ambient air  $^7\text{Be}$  activity is affected by climatic phenomena and that the impact of different climatic phenomena on  $^7\text{Be}$  activity is sensitive to location.

This is the first approach of determining the data and further analysis is needed for more accurate conclusions.

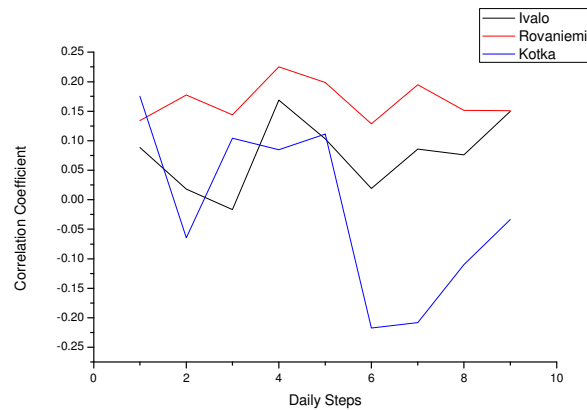


Figure 1. Day lag plot for the three locations in Finland.

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# **$^7\text{Be}$ activity concentrations on surface aerosols in Spain – Identification and meteorological analysis of peak periods**

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**Keywords:**  $^7\text{Be}$ , Spain, spatial distribution, synoptic meteorology

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The behaviour of  $^7\text{Be}$  activity concentrations have previously been analysed in several individual sampling stations in Spain. To complement the knowledge learned in these previous studies, a comprehensive analysis of its overall distribution in Spain is addressed in this work. Therefore, the current study analysed the behaviour of  $^7\text{Be}$  on surface air in Spain taking as reference four sampling stations included in the sparse monitoring network of the Radioactivity Environmental Monitoring (REM) database (<http://rem.jrc.ec.europa.eu/>).

The use of four sampling stations (Barcelona (41.38 N; 2.12 E), Bilbao (43.17 N; 2.94 W), Madrid (40.45 N; 3.69 W), and Sevilla (37.39 N; 6.01 W)), with different latitudinal location, as well as the relatively long time period used (2001-2010), allows to improve the understanding of its spatio-temporal distribution in southern Europe. In all of these stations, the  $^7\text{Be}$  measurements have a weekly temporal resolution but with different sampling interval (starting and ending dates) and the measurements were made on air filters collected at sampling networks (Hernández-Ceballos et al., 2015).

Figure 1 displays the temporal evolution of  $^7\text{Be}$  activity concentrations in each sampling station from January 2001 to December 2010.

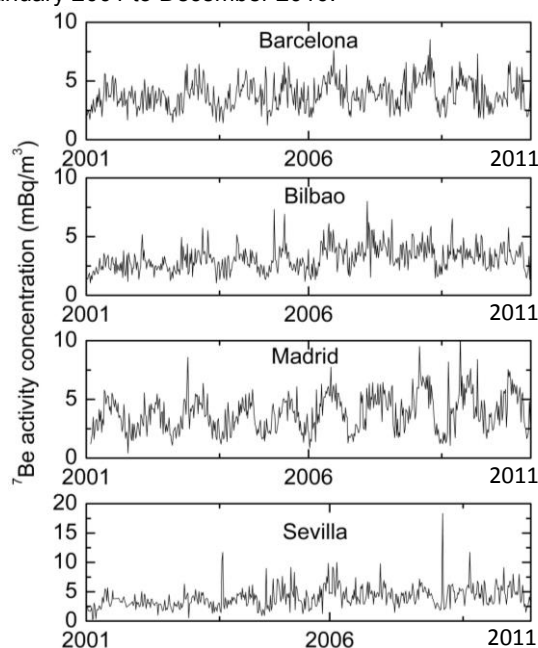


Figure 1.  $^7\text{Be}$  time series at Barcelona, Bilbao, Madrid and Sevilla from 2001 to 2010 (different scales).

The temporal evolution is very similar, with a sinusoidal behaviour (maximum values in spring and summer, and minimum values in autumn and winter). It is pointed out how the yearly size (max-min) of the cycle is reduced from north to south. The arithmetic mean for  $^7\text{Be}$  activity concentrations considering all the data available in each station ranges from  $3.1 \pm 1.1$  mBq/m<sup>3</sup> in Bilbao to  $4.0 \pm 1.8$  mBq/m<sup>3</sup> in Sevilla. The maximum values (above 90<sup>th</sup> percentile) are registered in Sevilla (6.16 mBq/m<sup>3</sup>), while the minimum ones (below 10<sup>th</sup> percentile) are registered in Barcelona (2.28 mBq/m<sup>3</sup>). This spatial distribution of the  $^7\text{Be}$  (increasing from north to south) marks the latitudinal impact on  $^7\text{Be}$  activity concentrations.

In order to assess the relationship between stations, the correlation coefficient (R) between time series of monthly values from 2001 to 2011 is successively calculated. A positive correlation between them is obtained, with R values always above 0.63, registering the maximum between Barcelona and Madrid (R = 0.81) and the minimum between Bilbao and Sevilla (R = 0.63). These results suggest a geographical division between the north and the south of Spain in terms of  $^7\text{Be}$  activity concentrations.

To analyse in detail this possibility, we have identified peak episodes of  $^7\text{Be}$  in Spain. With this aim, we first identify local episodes recorded at each sampling station (values above P90 from 2001-2010), followed by regional episodes for the whole region (coincidence of at least 3 (of 4) monitoring sites). The spatial distribution of these periods confirms the division of Spain in two geographical areas in terms of  $^7\text{Be}$  activity concentrations.

Once identified the regional episodes, we have analysed the synoptic meteorological conditions associated with each one with the aim to define the origin of  $^7\text{Be}$  and the set of meteorological scenarios in favour of high  $^7\text{Be}$  activity concentrations in Spain.

## **Acknowledgments**

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# Ambient dose equivalent rate response of gamma spectrometry monitors in the framework of MetroERM European project

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**Keywords:** *LaBr<sub>3</sub>, CdZnTe, ambient dose equivalent rate, angular response*

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In June 2014 started the European project "Metrology for radiological early warning networks in Europe" (MetroERM) with a period of three years (<http://earlywarning-emrp.eu/>). The aim of the project is focused to improve the metrological foundation of measurements (devices and methods) for monitoring airborne radioactivity and to introduce pan-European harmonisation in data reliability for area dose rate measurements which are input to the European Radiological Data Exchange Platform (EURDEP) and other monitoring networks.

Work Package 1 (WP1) of MetroERM aims to improve the harmonization of the measured ambient dose equivalent rate (ADER) data with emphasis on the next instruments generation, which will be based on compact spectrometers. Therefore, one task defined in this WP consists on validating these novel gamma spectrometry instruments by benchmarks experiments in calibration facilities and with Monte Carlo simulations. One of the selected facilities for the study is the Calibration and Dosimetry Laboratory (LCD) from the Institute of Energy Technologies (INTE) of the UPC. This laboratory is accredited by the Spanish National Accreditation Agency (ENAC), according to the ISO 17025 standard.

In a previous task of the MetroERM project two spectrometric monitors were selected to be characterized. These monitors are the SpectroTRACER monitor from Saphymo Company with a 1.5"x1.5" LaBr<sub>3</sub>(Ce) detector and the GS08x monitor designed by the Federal Office for Radiation Protection (BfS), which includes the compact GR1 module from Kromek Company. The GR1 incorporates a 1cm<sup>3</sup> CdZnTe detector, electronics and a Multichannel Analyzer (MCA). In addition, a 1"x1" LaBr<sub>3</sub>(Ce) detector from Saint-Gobain with a digital MultiChannel Analyzer (GMCA) developed by X-Ray imaging Europe Company, was also included in the study.



Figure 1. LCD facility with the SpectroTRACER monitor installed to be irradiated.

The monitors are being irradiated in the LCD facility to different ambient dose equivalent rates (ADER) and irradiation angles for different radionuclides (<sup>137</sup>Cs, <sup>60</sup>Co and <sup>241</sup>Am). In figure 1, the LCD facility with the SpectroTRACER monitor is shown.

For <sup>137</sup>Cs source the selected ADER irradiations are 2, 10, 50, 200, 2000, 5000 and 10000 µSv/h. For <sup>60</sup>Co, ADER values are 3, 10, 50 and 200 µSv/h. Finally, for <sup>241</sup>Am only 0.7 µSv/h is available for the study.

In order to study the angular response, the monitors are being irradiated at 0°, 30°, 45°, 60°, 75°, 90°, -30°, -45°, -60°, -75°, -90° incident angles according to the left side image of figure 2.

Due to the non-symmetrical geometry of the CdZnTe detector (1x1x1 cm<sup>3</sup>), the azimuthal angle dependence is also being studied for the GS08x monitor. Irradiations are being performed at 0°, 15°, 30°, 45°, 60°, 75°, 90°, 180° and 270° as shown in the right side image of figure 2

Regarding angular studies, the monitors are being irradiated to <sup>137</sup>Cs, <sup>60</sup>Co and <sup>241</sup>Am with ADER's in the range from 0.7 µSv/h to 10 µSv/h, depending on the source activity and the monitor efficiency.

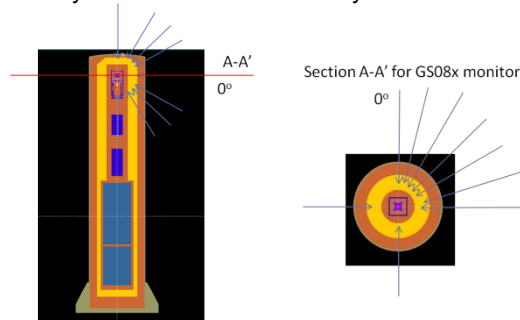


Figure 2. Polar (left image) and azimuthal (right image) angles used for the angular study. The geometries have been reproduced using the tools implemented in the PENELOPE Monte Carlo code.

The response of the spectrometric monitors to different gamma energies, ADER intensities and incident angles will be analyzed and presented. The results will be complemented with Monte Carlo simulations using PENELOPE/penEasy code.

## Acknowledgments

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# **<sup>7</sup>Be concentration in air surface over a long period of monitoring in the city of São Paulo, Brazil**

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Beryllium-7 ( $T_{1/2} = 53.3$  days), a cosmogenic radionuclide produced continuously in the upper atmosphere by cosmic ray spallation reactions with oxygen and nitrogen can be found in different compartments of the environment and its concentration is influenced by several factors, such as, temperature, precipitation, air velocity, air masses, altitude as well as latitude. It is quickly attached to aerosols after its formation, becoming a useful tool to study the dynamics of aerosol particles in the atmosphere. Long-term measurements of the spatial and temporal variation of <sup>7</sup>Be allow assessment of the influence of these factors (Valles et al, 2009).

Its subsequent deposition on the land surface, therefore, occurs as both wet and dry fallout, although it has been demonstrated that <sup>7</sup>Be fallout is primarily associated with precipitation. About 70% of the <sup>7</sup>Be is produced in the stratosphere and the remaining 30% in the troposphere. The amount produced in the stratosphere usually stays there for about 1 year before entering the troposphere, where it remains for about 6 weeks. Its transfer to the earth's surface is achieved by gravitational settling and precipitation (Lozano et al, 2011).

Cosmic radionuclide concentration in air at ground level can vary greatly according to latitude and altitude and generally the values measured at middle latitudes are higher than those at northern latitude (Cannizzaro et al, 2004). The production rate of <sup>7</sup>Be changes according to variations in flux of cosmic rays caused by the 11-year sunspot cycle. This phenomenon is one of the major parameters that cause the variation in <sup>7</sup>Be concentration in surface air (Papastefanou and Ioannidou, 2004).

Data of <sup>7</sup>Be concentrations in rainfall, air surface, soil and sediments is very well reported in the Northern Hemisphere; however these same results are limited in the Southern Hemisphere. Therefore, the aim of this work is to present results of the <sup>7</sup>Be concentration in air surface at the city of São Paulo, Brazil over a long period of time, from October 2001 to December 2014.

During this period <sup>7</sup>Be concentration was measured every 15 days at Instituto de Pesquisas Energéticas e Nucleares (IPEN), São Paulo, Brazil. The IPEN campus is located approximately 10 km west from downtown of the city of São Paulo, which is situated on a plateau in Southeastern Brazil, 23°33'58.27"S and 46°44'14.82"W and an average altitude of 760m above sea level. The climate in the area is temperate tropical with dry period in winter and rainy in summer. The

annual average temperature is 21.5°C, showing minimum and maximum of 12°C and 31.4°C, respectively. The annual rainfall in the city, for the studied period, averaged from 921 mm to 2088 mm.

In the framework of an environmental radiological monitoring program carried out at IPEN facilities, due to their routine gaseous effluents releases, atmospheric air is sampled at 1.0 m above the ground. The air is continuously pumped by a high volume sampler and forced to pass through cellulose filters with 47 mm diameter with retention efficiency near 100%, for the collection of particulates. The air volume sampled every 15 days corresponds to a volume about 3,000 m<sup>3</sup>.

Measurement of <sup>7</sup>Be was carried out by non-destructive  $\gamma$ -ray spectrometry using the 477.61 keV  $\gamma$ -ray. A coaxial Be-layer HPGe detector with 25% relative efficiency, 2.09 keV resolution at 1.33 MeV and associated electronic devices were used, with live counting time ranged from 100,000 to 300,000 s. The spectra were acquired by multichannel analyser Ethernim and, for the analysis, WinnerGamma software was used. The minimum detectable activity limits (MDA) was 0.31 mBq m<sup>-3</sup>. The activity was corrected for decay of <sup>7</sup>Be to the date of collection. The associated uncertainty for one sigma confidence ranged from 10% to 30% from the obtained results.

The results obtained were correlated to seasons, rainfall, temperature and sunspot number. <sup>7</sup>Be concentration in air surface showed a good agreement with data from Northern hemisphere of similar latitude, in spite of the city of São Paulo being situated in low latitudes of Southern hemisphere. The concentrations clearly displayed seasonal variations with greater values in spring and summer time and with a higher amount of precipitation.

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## Tritium and radiocarbon in seawater offshore Fukushima

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Tritium and radiocarbon belong to radionuclides, which release rates during the Fukushima accident and their concentrations in seawater offshore Fukushima have not been studied yet in detail. It has been expected that their release rates during the Fukushima accident have been much lower than other radionuclides, e.g. <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs (Povinec, Hirose, and Aoyama, 2013).

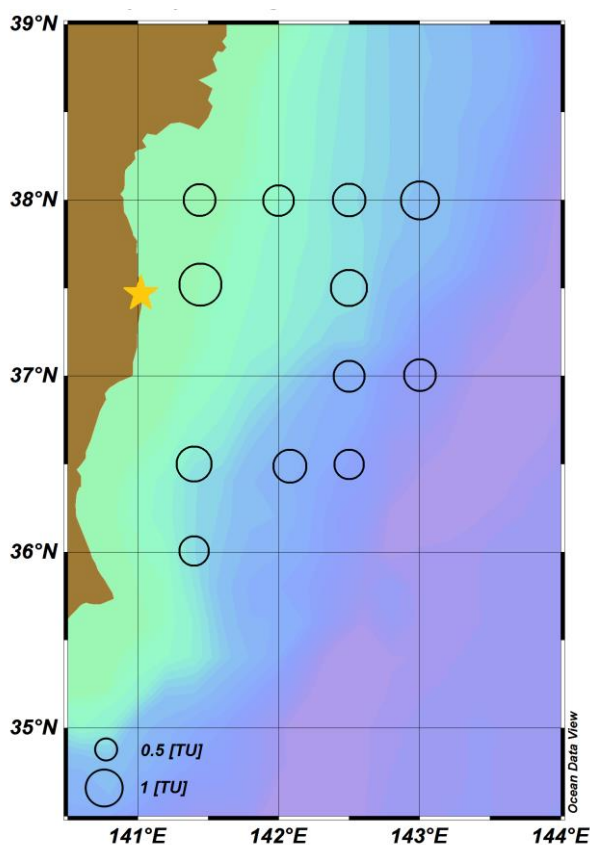


Figure 1. Distribution of <sup>3</sup>H in surface waters offshore Fukushima.

Tritium and radiocarbon levels in the marine environment will not pose therefore radioecological problems. However, the Fukushima releases have disturbed the well-established distribution of these radionuclides in the west Pacific Ocean, important for better understanding of processes in the water column, circulation of seawater masses and for climate change studies.

We shall present results of tritium and radiocarbon measurements in water column samples collected in June 2011 offshore Fukushima, and compare them with <sup>137</sup>Cs concentrations measurements (Povinec et al., 2013) in the same set of samples. While the levels of tritium and radiocarbon, measured at the sampling sites about 40 km from the coast, were a few times above the global fallout background from atmospheric tests of nuclear weapons (Povinec et al., 2005), the <sup>137</sup>Cs concentrations were by about three orders of magnitude above this background.

The water column data indicate that the transport of Fukushima-derived radionuclides downward to the depth of 300 m has already occurred. The observed radionuclide levels in surface waters (down to 600 km from the Fukushima coast) and in the water column (down to 400 m water depth) will be compared with predictions obtained from the ocean general circulation model (Nakano and Povinec, 2012).

### Acknowledgments

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# Beehive Model Creation for Use in Determining Radiation Dose to Bees and Bee Larvae

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**Abstract-** This research responds to the public concerns about radiation contamination to the environment. The objective is to develop a model of the honeybee hive using Monte Carlo N Particle (MCNP) computer code, and calculate absorbed fractions for multiple energies and multiple incident radiation types. This model could be of great value to the radiological community when being used in dose calculations to the environment, due to the critical role the honeybee plays in the ecosystem. Through their daily routines of collecting pollen, the honeybee may come into contact with radioactive material and subsequently contaminate its hive. Future work will examine a beehive located near a nuclear accident or test site, and determine the levels of contamination present. Additionally this research could be utilized to better understand the travel of nectar and pollen throughout the hive, aid in examining the insect partitioning of radionuclides, and speculate how much radiation deposited on a flower or pollen that bee collects.



# Study on the migration behaviour of Fukushima accident-derived iodine-129 from land area to the marine environment

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Keywords: <sup>129</sup>I, River system, Marine ecosystem, Migration behaviour, AMS.

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The March 2011 accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in Japan results in the massive release of high volatility fission products, including <sup>129</sup>I (8.1 GBq, Hou et al., 2013) and <sup>131</sup>I (120–160 PBq, Chino et al., 2011; Terada, 2012). The long-lived <sup>129</sup>I ( $T_{1/2} = 15.7$  million years) is one of the important radionuclide that we need to evaluate its migration behaviour from Japan's land area to the marine environment because of its relatively high chemical reactivity, biological concentration in the marine ecosystem, and affinity for the thyroid gland although it is less radiologically harmful than the short-lived <sup>131</sup>I ( $T_{1/2} = 8.02$  days). The present study aimed to evaluate the following three points: 1) the source and the discharge of particulate <sup>129</sup>I in the Niida River, 2) <sup>129</sup>I distribution in the marine sediment around estuary of the Niida River, and 3) <sup>129</sup>I distribution in the sea water and the marine organism in area close to the FDNPP.

In the Niida River system where the upstream are in the relatively high-contaminated area located 30–40 km northwest from the FDNPP, total suspended solid (SS) each month from December 2012 to January 2014 were continuously collected at Haramachi site (5.5 km upstream from the river mouth) of the downstream river by installing the time-integrative SS sampler. Marine surface sediments (0–3 cm depth) were collected at the two sites within 2 km from mouth of the Niida River. On the other hand, sea water samples of 2 L in water depths of 0 m, 5 m, 10 m, 15 m, and 20 m, and rock fish were collected at a site 6 km south-southeast of the FDNPP in July 2014. The iodine in SS, sediment, and rock fish of 0.2–0.5 g was volatilized and trapped in an organic alkaline solution (~10 ml) by pyrohydrolysis method (Muramatsu et al., 2008). After adding 2 mg iodine carrier to the trap solution and the filtered water sample of 1 L, the iodine was isolated and precipitated as AgI. The <sup>129</sup>I/<sup>127</sup>I ratio of AgI targets were measured using an AMS system at the Micro Analysis Laboratory Tandem Accelerator (MALT), The University of Tokyo (Matsuzaki et al., 2007). <sup>127</sup>I in the trap solution and the water sample were measured by an ICP-MS at the University of Tsukuba. The original <sup>129</sup>I activities and <sup>129</sup>I/<sup>127</sup>I ratios in the samples were calculated using <sup>127</sup>I concentration obtained from ICP-MS and <sup>129</sup>I/<sup>127</sup>I ratio obtained from AMS.

<sup>129</sup>I activities and <sup>129</sup>I/<sup>127</sup>I ratios in SS of the Niida River were 0.9–4.1 mBq kg<sup>-1</sup> and  $(2.5\text{--}4.4) \times 10^{-8}$ , which were strongly correlated with the total dry weight of monthly SS samples with correlation coefficient ( $R^2$ ) of over 0.79. <sup>129</sup>I activity and <sup>129</sup>I/<sup>127</sup>I ratio in SS were considered to infer the source of SS (relatively high-level contaminated upstream area or low-level contaminated downstream area). Meanwhile, the discharged particulate <sup>129</sup>I at Haramachi site were estimated to be

7.7–12 kBq month<sup>-1</sup> from September to October 2013. Therefore, relatively massive amount of particulate <sup>129</sup>I from the upstream were transported to the downstream of the Niida River from September to October 2013. <sup>129</sup>I activities in the surface marine sediments from two sites within 2 km from mouth of the Niida River were 5.8–8.4 μBq kg<sup>-1</sup>, 2–3 orders of magnitude lower than that of SS at Haramachi site of the Niida River.

The <sup>129</sup>I activity in surface sea water 40 months after the accident was 2.2 μBq L<sup>-1</sup> (Fig.1), 2 times larger than the previous <sup>129</sup>I data in surface sea water 3 months after the accident (Hou et al., 2013). <sup>129</sup>I activities in the rock fish were 42–48 μBq kg<sup>-1</sup>, approximately 20–400 times larger than that of sea water of same site. There is a possibility of biological <sup>129</sup>I concentration in the rock fish. Further investigations are needed to evaluate the mechanism.

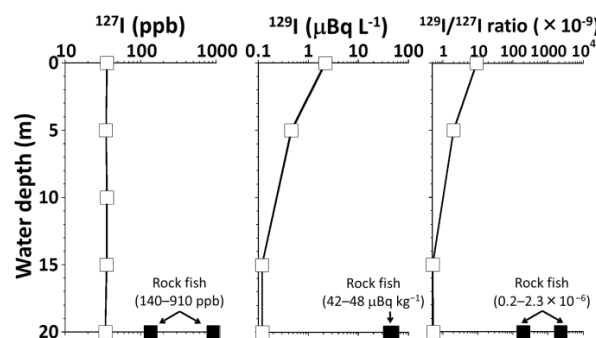


Figure 1 Profiles of <sup>127</sup>I concentration, <sup>129</sup>I activity, and <sup>129</sup>I/<sup>127</sup>I ratio in sea water (white squares) and rock fish (black squares) collected 6 km south-southeast of the FDNPP.

## Acknowledgments

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# Tritium level in the environment at Cernavoda Nuclear Power Plant after 15 years of operation

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**Keywords:** tritium, environment, CANDU reactor.

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The only Romanian nuclear power plant is Cernavoda Nuclear Power Plant (NPP). It is a Canadian Deuterium Uranium (CANDU) power reactor that uses heavy water reactor (deuterium oxide) as moderator and cooling agent, and natural uranium for fresh fuel. Cernavoda NPP has two nuclear reactors, first operating since 1996 and the other since 2007.

Tritium is the radioactive form of hydrogen and is a by-product of energy production in CANDU reactor. The release of this radioisotope in the environment is carefully managed by Cernavoda NPP with the respect of Romanian laws and exploitation licences. Taking into account more than ten years of nuclear activity on NPP site, a study of environmental tritium levels was conducted during 2009-2011 period.

Long term records from normal operation of Cernavoda Unit 1, wind pattern, meteorological conditions, and source terms data were used to evaluate areas of interest for environmental impact, conducting to a circle of around 30 km radius around NPP. Air circulation is concentrated mainly along the Danube Valley on the N-S direction. There were established 13 sampling locations situated at various distances from the source, from 1 km to 30 km, located mainly along Danube right bank. Tritium level evaluation was based on three years sampling campaigns during the spring and autumn. Different types of samples were collected from undisturbed biotopes by human activity.

Tritium measurements were performed for air, water, 0-10 cm, and 20-30 cm soil layers. A special attention was paid to wild vegetation present in each sampling location. Tissue Free Water Tritium (TFWT) was measured for herbaceous vegetation, swamp vegetation and trees leaves. The values shown below are the average value of tritium concentration measured in each sampling campaign for each type of samples. In case a sample was not collected (sampling campaign of September 2011 with a prolonged drought) this was not taken into consideration.

Water and sediment show similar behavior. Higher value for the sampling point located under 1 km radius was observed in Cismelei Valley (average below 40 Bq/l). The average value of tritium concentration in water established for the 12 sampling locations (without sampling location of exclusion zone of 1 km) was 2.42 +/- 0.43 Bq/l. It should be noted that no value exceeds the value of 100 Bq/l tritium concentration, accepted for drinking water (EC, Council Directive 96/83, 1998). Tritium concentration in sediment has an average of 1.04 +/- 0.24 Bq/kg fresh weight, fw, (without Cismelei Valley), maximum value of 29.76 +/- 0.46 Bq/kg fw being

recorded again in sampling location situated in exclusion zone.

For average values of tritium in air it is observed a descending trend depending on the distance from the source which is expected considering the dispersion of gaseous emission in the atmosphere. Maximum average value is recorded again in Cismelei Valley, 7.56 +/- 0.56 Bq/m<sup>3</sup>, established average value for the area of 30 km is 0.74 +/- 0.12 Bq/m<sup>3</sup> (without Cismelei Valley).

The same descending trend, depending on the distance, is also recorded for the average concentration of tritium in soil, with a maximum of 22.44 +/- 0.24 Bq/kg fw in the surface soil from Cismelei Valley. The established average value of monitored area was 1.33 +/- 0.12 Bq/kg fw (without Cismelei Valley). The aboveground soil (0-10 cm) has constantly showed higher values compared to the 20-30 cm soil layer which is influenced both by the exchange with the atmosphere and the precipitation's area.

If water, air and soil can be influenced by climatic conditions, and their values can be only instantaneous values, vegetation is the one that is totally influenced by the environment where it grows. The average value of tritium in water extracted from vegetation in the 30 km area (without 1 km exclusion zone) was 7.08 +/- 0.48 Bq/kg fw for all species of vegetation analyzed. Cismelei Valley presented higher values between 22.55 +/- 0.62 Bq/kg fw (Autumn 2010) and 882.22 +/- 1.03 Bq/kg fw (Spring 2011).

To conclude, regarding the behavior of tritium emitted through liquid and gaseous effluents by Cernavoda NPP, the only location in the 30 km radius which showed values higher than the level normally found in the environment is Cismelei Valley, sampling location situated in the exclusion zone of 1 km from NPP. Even so the recorded values for this location are still lower than the calculated values of tritium concentration below which there is no effect in the environment (Chouhan et al, 2009).

## Acknowledgments

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# Tritium level evolution in the environment at ICIT after 5 years of monitoring programme

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The only nuclear power plant (NPP) in Romania is Cernavoda NPP. It is a Canadian designed power reactor, with heavy water moderator and natural uranium fuel. Tritium is produced continuously in heavy water by neutron activation of deuterium present in high concentration. Taking into account the long period of operation of one unit of Cernavoda NPP, there was a clear need to develop the technology for tritium and deuterium separation from heavy water.

The Experimental Pilot Plant for Tritium and Deuterium Separation (PESTD) is an experimental installation within the national nuclear energy research program. The developed technologies use a combined liquid-phase catalytic exchange (LPCE) and cryogenic distillation in order to remove tritium from CANDU moderator, heavy water. Total radioactive source of PESTD is contained in the installation and it is equal with tritium inventory of process fluids. Calculated derived release limits are: for gaseous effluents  $2.5 \times 10^{13}$  Bq/year (tritiated vapours),  $6.04 \times 10^{14}$  Bq/year (tritium gas), and for liquid effluents  $7.98 \times 10^{12}$  Bq/year (tritiated water). Until now, the processes described above were tested using heavy water and tritiated water below exemption level of the Romanian national legislation (CNCAN, 2000).

PESTD is part of the Institute for Cryogenics and Isotopic Technologies (ICIT), situated at a distance of about 10 km from the city Rm. Valcea. It is close to several rural settlements, and it is located on the city's industrial estate. All wells near the industrial estate are out of use, and the drinking water is supplied from a groundwater source at a distance of about 9 km from ICIT. The Olt River is the only source for industrial water supply and is situated about 1 km from ICIT. The wastewater is discharged outside the Institute in the sewage system of industrial estate.

There were established five locations considering wind patterns, meteorological conditions, and source data terms, Figure 1. The population group most exposed to radiation due to the operation of PESTD is the ICIT personnel not working in the experimental installation. Three of the five locations are inside the Institute's yard. The other two locations are situated at 2 km and 7 km on S direction, and they are used as control locations for gaseous and liquid effluents.

Tritium measurements were performed in air, surface water, and precipitation on a monthly basis. The method used was liquid scintillation counting. The soil and sediments were measured once per year. The home-grown fruits, vegetable and grains from the gardens located in the vicinity of PESTD were collected

monthly during the growing period, each year. Tissue Free Water Tritium was measured using azeotropic distillation and liquid scintillation method.

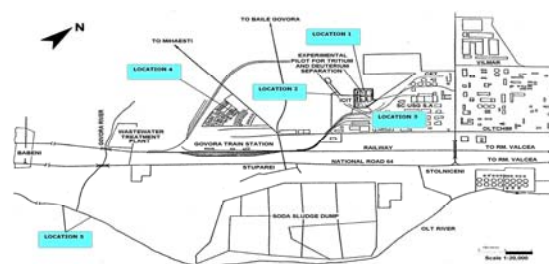


Figure 1 Site and sampling locations for PESTD

Tritium concentration in precipitation during the monitoring period, 2010-2014, exhibits an average of  $10.8 \pm 1.7$  TU (1 TU = one tritium atom to  $10^{18}$  hydrogen atoms). A minimum value below 4.5 TU in November 2014 and a maximum of  $21.9 \pm 2.1$  TU in July 2014 have been recorded. These are typical values that can be found in precipitation for Vienna, monitoring station of Global Network of Isotopes in Precipitation.

The yearly average tritium concentration for the five locations varied between  $11.6 \pm 2.52$  mBq/m<sup>3</sup> in 2013 and  $23.2 \pm 3.39$  mBq/m<sup>3</sup> in 2012. A minimum value of  $3.8 \pm 3.39$  mBq/m<sup>3</sup> has been recorded in December 2013, and a maximum of  $97 \pm 3.39$  mBq/m<sup>3</sup> in February 2012. These values are slightly higher than recorded values for the preoperational monitoring programme (Varlam et al, 2011).

The values of tritium concentration in surface water, soil, fresh products used in human consumption, didn't exceed 4 Bq/kg. Nuclear activity of PESTD didn't have any impact in the environment until now.

## Acknowledgments

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# INVESTIGATING RADIOLOGICAL SITUATION FEATURES AT P-1 TECHNICAL SITE

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“Experimental Field” was the first testing ground of the Semipalatinsk Test Site and was designed for atmospheric (ground and air) nuclear tests performed between 1949 and 1962. There was a series of technical sites (P-1, P-2, P-3, P-5, P-7) within “Experimental Field” site where ground nuclear tests were performed. P-1 technical site which is located in the center of “Experimental Field” was the object of research. According to the available historical data (USSR chronology of nuclear tests is 1949-1962) 4 ground nuclear tests have been performed in the P-1 area (29.08.1949 – 22 kt, 24.09.1951 – 38 kt, 12.08.1953 – 400 kt and 05.11.1962 – 0.4 kt).

P-1 technical site represents a plain with insignificant altitude difference and considerable anthropogenic activity trace. The outline of a round shape area has a radius of 2 km and an area of 13 km<sup>2</sup>. Strong test infrastructure was constructed at this site in the form of various ground and underground concrete structures, caponiers, cable channels, other anthropogenic objects designed for registering basic nuclear explosion parameters. Currently the P-1 technical site center has an asymmetric crater with signs of groundwater as well as a variety of artificial objects as earth embankments.

The main purpose of research was obtaining detailed information of the present state of radionuclide contamination in the P-1 technical site area as well as of its forming features.

Investigation of areal radionuclide distribution features consisted in obtaining up-to-date information on spatial distribution of artificial radionuclides around the site area by applying pedestrian gamma-spectrometric survey and analyzing soil samples using gamma-spectrometric and radiochemical methods under laboratory conditions. The applied methods allowed to obtain data from which test venue identification was carried out in the area of the object being investigated.

As a result of works performed, general nature of contamination in the site area was determined. Two epicenters of nuclear tests as well as fallout plumes have been discovered. The revealed epicenters have individual radionuclide picture and different isotopic ratios.

In the epicenter located in the center of the site, maximum specific activity for <sup>214</sup>Am does not exceed 3500 Bq/kg, <sup>137</sup>Cs values reach up to 80000 Bq/kg, mean <sup>241</sup>Am/<sup>137</sup>Cs ratio value is 0.04, values for <sup>238+240</sup>Pu/Am isotopic ratios vary from 3 to 70. Taking into account findings and literary data one can assume that it is exactly the point where 3 tests were performed (29.08.1949 – 22 kt, 24.09.1951 – 38 kt, 12.08.1953 – 400 kt). The diameter of epicentral zone is ~2000 meters and is related to the test performed in 1953.

A second epicenter is located at a distance of ~1200 meters to the center of P-1 technical site. Maximum specific activity value for <sup>214</sup>Am is about 80000 Bq/kg, for <sup>137</sup>Cs – 7000 Bq/kg, The mean <sup>241</sup>Am/<sup>137</sup>Cs ratio value is 11.3, for <sup>239+240</sup>Pu/<sup>241</sup>Am – 6.8, which in turn indicates belonging to a separate experiment. The diameter of the epicentral zone is ~600 meters.

Application of data on <sup>152</sup>Eu (about  $n \cdot 10^4$ ) was found to be a crucial moment which allowed to identify tests epicenters and to characterize the dimensions of epicentral zones.

Upon the results of work the information has been obtained on present radiation condition at P-1 technical site formed in the area. The revealed epicenters have been identified, characterized and fully correspond to the list of tests performed.

# Spatial and vertical distribution of $^{137}\text{Cs}$ in a Greek area following the Chernobyl nuclear reactor accident

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Keywords:  $^{137}\text{Cs}$ , Chernobyl, Trikala, Greece.  
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## Introduction

The Prefecture of Trikala is one of the most contaminated areas in Greece after the accident at the Chernobyl nuclear power plant in 1986 (Petroopoulos *et al.*, 1996). Heavy rainfall along with the passage of radioactive cloud over Greece caused local highly radioactive deposits of  $^{137}\text{Cs}$  (Papastefanou *et al.*, 1988). However, there is no data concerning the fate of  $^{137}\text{Cs}$  in this area after 27 years of the nuclear plant accident. In this study, an effort has been undertaken to get field measurement data concerning the spatial and vertical distribution of the deposited  $^{137}\text{Cs}$  in the Prefecture of Trikala.

## Methods

**Sampling.** A number of soil core samples (~20) of about 30-60 cm length and 74 mm in diameter were taken from the study area during 2013, using a stainless steel corer of a length of 150 cm and a diameter of 3 cm. After sampling, each core was forced out of the corer and sliced into horizontal layers of 0-10 or 0-20 cm thickness. The first layer was used to examine the spatial distribution of  $^{137}\text{Cs}$ . A differential GPS with an RM accuracy of about  $\pm 1\text{m}$  was used for the positioning of each core location.

**Sample preparation.** The soil samples were grinded, and then passed through a 2 mm mesh screen to remove pebbles and plant debris. In order to remove moisture, the samples were placed in an oven at  $104^\circ\text{C}$  for 24h. Finally, all samples were transferred to standard counting cups, weighed and sealed tightly.

**Analyses.** The activity concentration measurements of  $^{137}\text{Cs}$  in the soil samples were performed by means of a high resolution  $\gamma$ -ray spectrometry set-up combined with a Canberra HPGe detector.

## Results

The spatial distribution of  $^{137}\text{Cs}$  activity concentration varied considerably and ranged between  $3.2$  and  $176\text{ Bq kg}^{-1}$ . These variations, even at very short distances, such as noticeable on samples of Trikala, depend primarily on rainfall at the time the plume passed over (Simopoulos, 1989).

The maximum depth of cesium occurrence was at 60 cm, and the greater fraction of its concentration was found, in most cases, in the upper layer. As concerns the vertical distribution of  $^{137}\text{Cs}$ , results from a number, but not of all of the examined cores, seems to fit reasonably well to an exponential function (Figure 1) (Letho *et al.*, 2013).

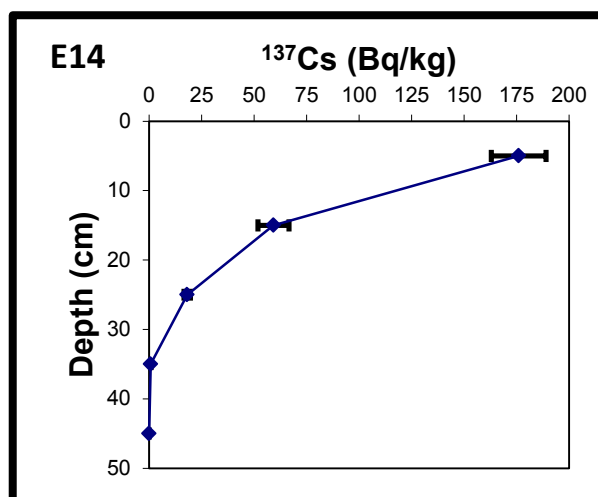


Figure 1. Depth distribution of  $^{137}\text{Cs}$  activity concentration.

The inventory of  $^{137}\text{Cs}$  was also calculated at a selected number of cores by summing the radioactivity over the whole depth and taking into account the decay over the 27 years. As shown in Table 1, the average value of inventory was  $18.6 \pm 4.17\text{ kBq m}^{-2}$ . This value is of the same order of magnitude compared to those of other researchers (Simopoulos, 1989).

Table 1. Inventory concentrations of  $^{137}\text{Cs}$ .

Sample Code	Depth (cm)	$^{137}\text{Cs}$ ( $\text{kBq m}^{-2}$ )
E12	0-60	$13.5 \pm 1.05$
E14	0-50	$18.8 \pm 1.67$
E16	0-60	$18.4 \pm 1.44$
E17	0-50	$23.7 \pm 1.68$
Average $^{137}\text{Cs}$		$18.6 \pm 4.17$

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Letho, J., Vaaramaa K. and Leskinen (2013). *J. Environ. Radioact.* **116**, 124-132.

# Estimation of air and surface radioactivity concentrations from pulse height distribution measured by NaI(Tl) scintillation detector

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To evaluate internal and external doses in a nuclear emergency, air and surface radioactivity concentrations are essential. However, it is extremely laborious to measure them as compared with gamma dose rate. The purpose of this study is, therefore, to establish a method for estimating the air and surface radioactivity concentrations of anthropogenic nuclides from pulse height distribution data routinely measured with NaI(Tl) scintillation detectors at monitoring stations and to apply it to the Fukushima Daiichi NPP accident case to test the method.

The principle of the method is to find activity concentrations of anthropogenic radionuclides which best reproduce observed pulse height distributions when they are used as input data to Monte Carlo photon transport calculations. In air concentration estimation, we used the method of our previous study (Hirouchi et al., 2014), which used the fact that the count rate of low energy part (<500keV) of pulse height distribution differs depending on whether nuclides are in air or on the ground surface.

The calculation of pulse height distribution was carried out with the Monte Carlo photon transport code EGS5 (Hirayama et al., 2005). Radionuclides were assumed to exist uniformly both in the atmosphere and on the ground surface. In this study, eight anthropogenic radionuclides (<sup>133</sup>Xe, <sup>131</sup>I, <sup>132</sup>I, <sup>133</sup>I, <sup>134</sup>Cs, <sup>136</sup>Cs, <sup>137</sup>Cs, <sup>132</sup>Te) were selected, which have high volatility, large potential contribution to dose rate, and were detected by NaI(Tl) scintillation detectors after FDNPP accident. <sup>133</sup>Xe was assumed to exist only in air. Eight energy ranges were set for the main gamma rays of the nuclides. The surface concentration was assumed to be proportional to that in air for each nuclides. The air to surface concentration ratio  $F$  was determined by the energy range 120-180keV, which consists only of scattered photons. Air and surface concentrations of the objective nuclides were determined with a least-square method in which the square sum of relative differences of the count rate in each energy range between observation and calculation was minimized. We have applied this estimation method to pulse height distribution measured in Ibaraki prefecture, where plumes from FDNPP were detected.

It was found that estimated surface concentration values of <sup>134</sup>Cs and <sup>137</sup>Cs were constant after March 21, and within a factor 2 of measured values. Temporal decrease of each estimated nuclide showed good agreement with its physical half-life. Air concentration of

each selected nuclide was estimated to be within a factor 3 of the measured values (Table 1).

The lower limit of concentration that can be estimated by this method is limited by interference from the coexisting natural radionuclides such as <sup>214</sup>Pb and <sup>214</sup>Bi and the statistical error in the count rates. The lower limits were evaluated to be 89, 71, and 51 Bq m<sup>-2</sup> for <sup>131</sup>I, <sup>137</sup>Cs, and <sup>134</sup>Cs, respectively. It is also to be noted that this estimation method cannot be applied to cases where dose rate exceeding about 15 μGy/h, because pulse height distributions become saturated.

It can be concluded from the results above that activity concentrations can reasonably be estimated from pulse height distributions routinely measured at monitoring stations. Estimated values showed good agreement with measured values. This result suggests that the method can quickly provide useful information in nuclear emergency.

Table 1. Comparison of estimated and measured air activity concentrations in Tokai, Ibaraki at 5 JST on 21 March, 2011 (Ohkura et al., 2012).

Nuclides	Air concentrations (Bq m <sup>-3</sup> )	
	estimated	measured
<sup>131</sup> I	1100	1920
<sup>132</sup> I	540	341
<sup>132</sup> Te	420	237
<sup>134</sup> Cs	180	426
<sup>136</sup> Cs	70	53
<sup>137</sup> Cs	170	426

## Acknowledgments

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# Atmospheric dispersion and surface deposition of radionuclides discharged by Fukushima Dai-ichi Nuclear Power Plant accident

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**Keywords:** Fukushima Dai-ichi Nuclear Power Plant accident, atmospheric dispersion model, surface deposition, WRF.

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It is essential to reveal the evolution of atmospheric dispersion and surface deposition of radionuclides discharged from Fukushima Dai-ichi Nuclear Power Plant (FDNPP) for estimating doses to the public during the initial stage of the accident, March 2011. Atmospheric transport models play important roles because the temporal and spatial coverage of the environmental monitoring data is sparse. In this study, we discussed the evolution of atmospheric dispersion and surface deposition by using the atmospheric dispersion model.

Hourly atmospheric concentrations and hourly surface depositions of discharged radionuclides were calculated by coupling the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) and a Lagrangian particle random-walk model (LPRM) (Hirao et al., 2013). WRF calculates the three dimensional meteorological fields. Advection, diffusion, radioactive decay and deposition (dry and wet) were calculated in the LPRM. The behaviour of <sup>131</sup>I and <sup>137</sup>Cs was calculated as passive tracers. The calculation domain was an 835-km square with a 10-km depth above the ground to cover most of Tohoku and Kanto regions. For initial and boundary conditions and the four-dimensional data assimilation of the meteorological fields, the mesoscale analysis (MANAL) from Japan Meteorological Agency was used. The model calculation was conducted for the period from 4 Local Time (LT), March 12, 2011 to 0 LT April 1, 2011. The source term was used from Terada et al. (2012).

The outline of the evolution of atmospheric dispersion and deposition calculated with the model is shown in Figure 1, and as follows; 1) On March 12, the radioactive plume, which has high atmospheric concentration due to a hydrogen explosion and ventilation at Unit 1, was transported to the northwest direction from FDNPP and caused a substantial dry deposition. 2) From the morning of March 15 to the noon, the radioactive plume was transported to southwest direction from FDNPP and caused a substantial amount of a dry and fog deposition in North Kanto. In the afternoon the plume changed its direction clockwise to the northwest of FDNPP and caused a significant amount of wet deposition at Fukushima that was main contamination in the accident. 3) On March 20, the radioactive plume was lifted and transported over Yamagata and Miyagi. 4) On March 21, the plume was transported to the Kanto region and caused a large amount of wet deposition in Ibaraki and Chiba. Until March 25, the plumes were transported several times over the Honshu Island. 5) On March 30, the plume was transported to Kanto region and resulted in a large amount of wet deposition in Fukushima.

The comparisons between observed hourly <sup>137</sup>Cs concentrations at 56 stations in Kanto regions (Tsuruta

et al. 2014) and calculated ones were also carried out during March 15-16, 20-23, 2011. It was found that 51% of calculated concentrations were within a factor of 5 of observed concentrations. The arrival times of the plume calculated with the model agreed with the observed ones in the range of  $\pm 3$  hours.

We concluded that the atmospheric dispersion processes and the present surface deposition distribution of <sup>137</sup>Cs over eastern Japan could be reasonably produced by the atmospheric dispersion model.

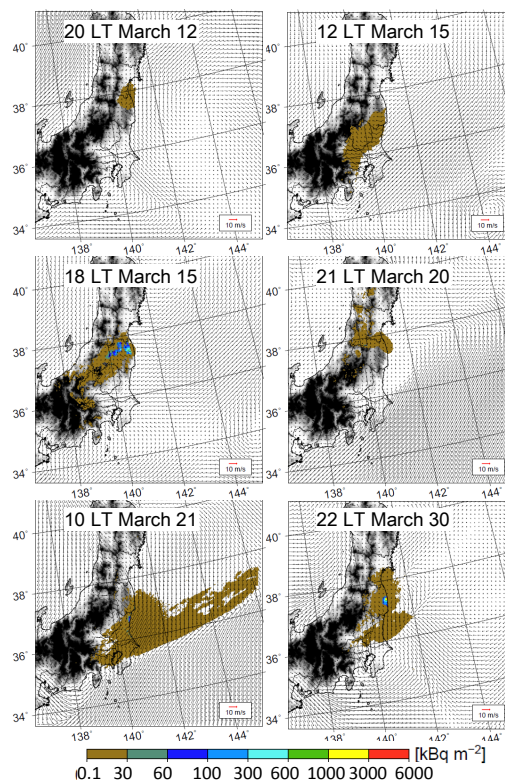


Figure 1. Calculated hourly surface deposition of <sup>137</sup>Cs.

## Acknowledgments

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# Long-term variations of atmospheric radionuclides in Bratislava air

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**Keywords:** atmosphere, radioactivity, radionuclide variations  
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Monitoring of atmospheric radionuclide concentrations at the Mlynska dolina locality (Bratislava) with week periodicity started from 2003. Aerosols containing  $^7\text{Be}$ ,  $^{40}\text{K}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  in the ground-level atmosphere were collected on nitro-cellulose filters, with the average flow rate of about  $80 \text{ m}^3/\text{hour}$ . After one week period, exposed filters were gamma-spectrometrically analyzed using HPGe detectors situated in low background shields (Sýkora et al. 2008).

Measured concentrations of  $^{210}\text{Pb}$  and  $^7\text{Be}$  ranged from  $0.20$  to  $3.01 \text{ mBq m}^{-3}$  with the mean value of  $0.63 \pm 0.03 \text{ mBq m}^{-3}$ , and from  $0.22$  to  $5.5 \text{ mBq m}^{-3}$  with the mean value of  $2.27 \pm 0.06 \text{ mBq m}^{-3}$ , respectively. Measured concentrations of  $^7\text{Be}$  are shown in Fig.1. Seasonal variations in concentration of both radionuclides were observed. Concentrations of  $^7\text{Be}$  show typical trends for inland country of the Northern Hemisphere with maxima in spring and early summer, and minima in winter. Concentrations of  $^{210}\text{Pb}$  (a radon decay product) are different and show minima in summer and maxima in winter. Seasonal variations of activity concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  show thus typical mutually inverse trends.

In order to measure low concentrations of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in atmospheric aerosols, monthly samples were accumulated from weekly sampled filters.  $^{137}\text{Cs}$  concentration shows decreasing trend (Sýkora et al, 2012). This radionuclide was introduced to the atmosphere by global fallout and the Chernobyl accident. Higher concentrations in winter months refer to resuspension of soil particles to the atmosphere. The aerosol component of the ground level atmosphere in Bratislava showed typical values of activity concentrations also for  $^{137}\text{Cs}$  and  $^{40}\text{K}$ , as expected for the Central Europe.

The activity concentration of anthropogenic  $^{137}\text{Cs}$  in ground level atmosphere is  $\sim 0.6 \text{ } \mu\text{Bq m}^{-3}$ . Mean activity concentration of primordial  $^{40}\text{K}$  in aerosols reaches values around  $5.0 \text{ } \mu\text{Bq m}^{-3}$ . The  $^7\text{Be}/^{210}\text{Pb}$  activity ratios are presented and correlation study has

been carried out between the meteorological factors and concentrations of radionuclides. The observed mean values can be considered as representative at ground-level air in our geographical region. The environmental radioactivity monitoring can be useful to study the transport processes between the troposphere and low-level atmosphere.

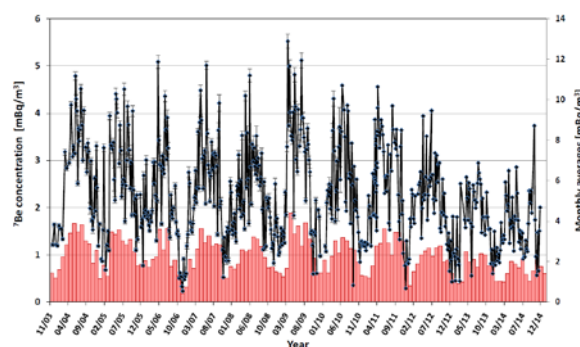


Figure 1. Time variation of  $^7\text{Be}$  aerosol concentration in the Bratislava atmosphere

## Acknowledgments

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# The valley system of the Jihlava river and Mohelno reservoir with enhanced tritium activities

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**Keywords:** tritium, NE-OBT, biota, nuclear power plants

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The Dukovany nuclear power plant (NPP Dukovany) releases liquid effluents, including HTO (tritiated water), into the Mohelno reservoir, located in a deep valley.

Significantly enhanced tritium activities were observed in the form of NE-OBT (not exchangeable organically bound tritium) in the surrounding biota which lacks direct contact with the water body. This indicates a possible tritium uptake by plants from air moisture and haze, which is, besides the uptake by roots from soil, one of the most important tritium transfer mechanisms from environment to plants (Baglan et al., 2011; Boyer et al., 2009). In this valley with limited possibilities of ventilation, observed tritium activities exceed by more than one order of magnitude the background reference level, which is below 2 Bq/L for corresponding HTO (Svetlik et al., 2014; Hanslik et al., 2009).

Quite an unusual effect occurs in the technological framework of this NPP, where a cooling water drawn from the Mohelno water reservoir is utilized. Liquid discharges from this NPP are released by outlet drainage canal into the Skryjský creek, which flows into the Mohelno reservoir about 50-60 meters far from the water inlet of the NPP. The short distance between these two places causes a "loop" effect and boosts activities of HTO at this place.

As we have found, the local land relief which determines the air exchange at the site probably has a dominant effect on the NE-OBT activities within this valley system.

Likewise, elevated tritium activities can also be found in the Dalešice reservoir which is several times greater than Mohelno and located upstream of it. The Dalešice lake harbours pump storage hydroelectric power plant, hence tritiated water from Mohelno occurs in this reservoir also.

Downstream the Mohelno reservoir, the effluent part of the Jihlava river runs on through this long deep valley.

In our presentation, we will describe the whole system with enhanced tritium activities found within this valley, and we will discuss both HTO and NE-

OBT occurrence in various parts of this interesting system.

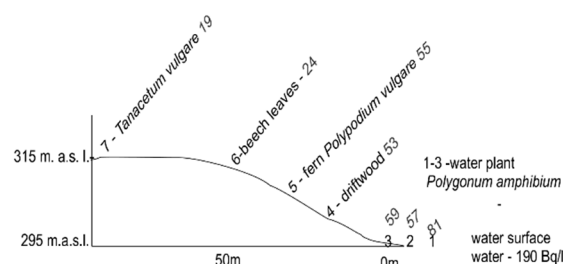


Figure 1. Schematic drawings showing the values of NE-OBT in biota samples from the Mohelno reservoir, 50 m of the mouth of the Skryjský creek (values are reported in Bq/kg of dry mass).

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# Development of rice reference material and a proficiency test for the measurement of Cs-137 and K-40

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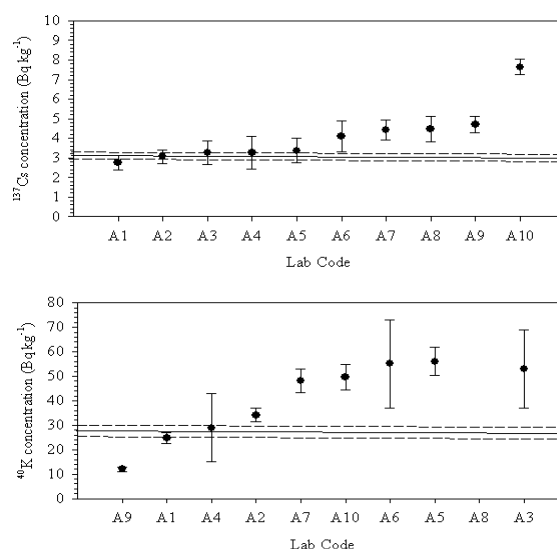
Keywords: Reference Material, Rice,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ . Intercomparison exercise  
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For a laboratory to produce consistently reliable data, it must implement an appropriate programme of quality-assurance and performance-monitoring procedures. Intercomparison exercise is one of these procedures. The usual format for Intercomparison exercise schemes is based on the distribution of samples of a test material to the participants. In accordance with the RCA (Regional Cooperative Agreement) /UNDP(United Nation Development programme) Project Work Plan, the international intercomparison exercise was organized and 10 Asian countries participated on the programme. The purposes of the intercomparison exercise were to provide the quality assurance on the data produced by the participating laboratories, to review performance-monitoring procedures and to exchange technical information.

The reference material used for the intercomparison exercise was rice material which was developed by KRISS for the measurement of Cs-137 and K-40.

To produce rice bearing Cs-137, KRISS was in collaboration with KAERI (Korea Atomic Energy Research Institute, Korea). The rice culture was conducted from the paddy field in the greenhouse located in KAERI. An aqueous carrier-free radioactive solution (74-93 kBq mL<sup>-1</sup> as  $^{137}\text{CsCl}$ ) was applied to distribute uniformly on the water surface and then mixed with the topsoil to about a 15 cm depth (Choi et al., 2005). As the quantity of material obtained from KAERI was small and the activity of Cs-137 was too high to carry out PT, the sample was mixed with natural rice ( $\text{Cs-137} < \text{MDA}$ ).

The homogeneity on the rice material was tested by measuring Cs-137 and K-40. The rice samples were stored in 4 bulk containers, and 2 samples (total 7) were taken from each bulk container and Cs-137 and K-40. The low-level background gamma-ray spectrometer (LB-HPGe) developed by KRISS (the relative efficiency 120 %) was employed to determine the reference activity for Cs-137 and K-40 in the rice sample. The integral background of counting rate of LB-HPGe detector was 1.61 s<sup>-1</sup> from 50 to 3000 keV and the count rate of  $^{40}\text{K}$  measured at 1461 keV was 1.01×10<sup>-3</sup> s<sup>-1</sup>. The relative uncertainties of homogeneity are estimated to be 2.2 % for Cs-137 and 3.5 % for K-40, respectively. The reference values and the participant's results for Cs-137 and K-40 showed at Fig. 1.



While the results of Cs-137 reported by participants were relatively concordant with the reference value, K-40 results were higher than the reference value. This is attributable to the difficulty of K-40 measurement due to the low concentration of K-40 in the rice reference material and a high background by contributing a natural K-40 radioactivity.

## Acknowledgments

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# Study of aerosol $^{137}\text{Cs}$ activities around nuclear power plants in Slovakia

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Keywords: nuclear power plant,  $^{137}\text{Cs}$ , atmosphere, Bratislava.

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Nuclear power plants (NPP) impact studies on the environment have been of many discussions during the last years, especially after the Fukushima accident. There are two NPPs under operation in Slovakia (central Europe), one in Jaslovské Bohunice and the second one in Mochovce. Both NPPs operate pressurized light water reactors of Russian origin (VVR-440) with 440 MW<sub>el</sub>. One of frequently studied radionuclides, which is produced in nuclear reactors and may be released to the environment is  $^{137}\text{Cs}$ . Most of the atmospheric  $^{137}\text{Cs}$  in this region comes, however, from the resuspension of the Chernobyl-derived  $^{137}\text{Cs}$ , as well as caesium produced during nuclear weapons testing, mainly during the 1960's (Kulan, 2006).

Atmospheric aerosol radioactivity has been continuously monitored at both NPPs. There is also a long tradition in monitoring of radioactivity in the Bratislava air by the Department of Nuclear Physics and Biophysics of the Comenius University. The first measurements of  $^{137}\text{Cs}$  on aerosols were carried out since 1976 (Povinec et al., 2012).

The objective of this work has been to study local trends and variations of anthropogenic  $^{137}\text{Cs}$  in the troposphere at each NPP location, to estimate a radiation impact of the NPPs on the local environment, and to compare the obtained results with the Bratislava monitoring station.

Atmospheric aerosols have been collected in weekly/monthly intervals on filters by pumping of large volumes of air (at least 50 000 m<sup>3</sup> per month). In most cases the used detection system, low-level gamma-ray spectrometers with HPGe detectors, reached only detection limits for weekly samples, therefore monthly intervals were chosen for the determination of  $^{137}\text{Cs}$  activities (Šýkora et al, 2008).

A typical trend in the  $^{137}\text{Cs}$  activity concentration in the air can be seen on Figure 1. In Bratislava aerosols, local maxima of concentrations are clearly distinguishable in winter seasons. Similar trends can be observed also in  $^{137}\text{Cs}$  concentrations at the Mochovce monitoring station. These maxima are connected with increased resuspension of particles from the soil due to enhanced windy conditions during the winter seasons.

Activity concentrations of  $^{137}\text{Cs}$  in Mochovce and Bratislava stations are approximately at the same level.

Such trend is not clearly visible in  $^{137}\text{Cs}$  concentrations measured at the Jaslovské Bohunice station, where the activities are higher in comparison to other locations. This increase of  $^{137}\text{Cs}$  in the atmosphere is connected with decommissioning of A-1 reactor after accident in 1977 (INES 4).

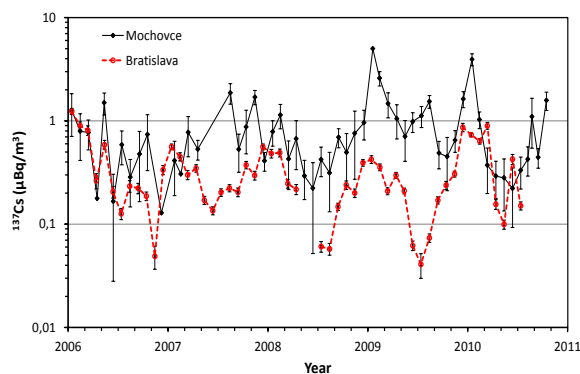


Figure 1. Comparison of monthly  $^{137}\text{Cs}$  concentrations in Bratislava and near the Mochovce NPP.

## Acknowledgments

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# Application of the atmospheric dispersion modelling in the intermediate and late phase of nuclear accident

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Keywords: radiation, meteorology data, long-term measures, emission  
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Determination of the environmental radiation conditions is very important in the course of nuclear or radiological accidents. Based on these conditions the projected doses are determined and various countermeasures can be taken.

Decision support systems are given for early, intermediate and late phases of nuclear and radiation accidents. In the intermediate and late phase radioactive materials are not probable to be released into the environment.

The urgent and early phases exist from observation of accident to the end of major environmental emission. In these phases the source term and the meteorology are not known but can be predicted.

In the intermediate phase there is no primary emission. In the late phase there is no emission any longer but the deposition and dispersion of radioactive materials persist.

In order to provide an adequate dose consequence for areas affected by the emission it is necessary to assess the impact and distribution of radioactivity in the environment.

The spatial distribution of the environmental contamination can be estimated by an appropriate atmospheric dispersion program, e.g. SINAC [1] (developed in MTA Centre for Energy Research) or RODOS (developed in international co-operation). As part of emergency preparedness activities various emergency planning zones and distances should be designated around the reactor [2]

In the intermediate and late phases the source term and the meteorology situation during the dispersion are known and some monitoring data are available e.g. dose rates and results of measurements of in situ gamma-spectrometry. However these data are limited in space and in time.

Knowing the meteorological situation the dispersion of the atmospheric contamination can be described. The most important meteorological data are the following: wind speed, wind direction, precipitation, atmospheric stability and height of atmospheric boundary layers. These data are available in several ways:

1.) The forecast of numerical weather models like AROME, ECMWF, WRF. The forecast file of AROME (applied by the Hungarian Meteorological Service) is available in every six hours (00, 06, 12, 18 UTC) up to 36 hours. The most accurate information can be obtained by using the first six hours from every single forecast file.

2.) The model can be run in the intermediate time (03, 09, 15, 21 UTC). Therefore more accurate results can be obtained than for the 6 hours forecast.

3.) Measured data used for the prediction of the meteorological situation are available in the late phase of the nuclear accident and these data can be used to specify the meteorological input.

4.) The meteorological inputs can be refined with the various additional measurements, SODAR, soundings (radiosonde), measurements on drones.

5.) The doses from the radioactive contamination are strongly influenced by the precipitation. The meteorological RADAR system describes the most accurately the precipitation field.

Introduction of (further) countermeasures should be based on radiological measurements in the intermediate and late phases, however, accurate atmospheric dispersion modelling can help improve and broaden the measurement database.

Proposed methods of decision support in the late phase of a nuclear accident based on atmospheric decision modelling will be presented.

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[2] IAEA: EPR NPP Public Protective Actions 2013.



# Laboratory infrastructure for radon and thoron measurements by CDs/DVDs

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**Keywords:** radon, thoron, CD, laboratory infrastructure.

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Recent reports raised concern about the possible impact on indoor radon of two modern technologies: fracking technology for natural gas production (Casey et al., 2015) and energy-efficiency building interventions (Milner et al., 2014). One method that shows capability to address this new challenge in environmental radioactivity studies is that based on analysis of CDs/DVDs stored indoors. The method allows retrospective measurements of radon ( $^{222}\text{Rn}$ ) (Pressyanov et al., 2001; 2010) and thoron ( $^{220}\text{Rn}$ ) (Pressyanov 2012), including detection of significant changes in the concentrations after technological interventions in the past (Pressyanov et al., 2015).

The CD/DVD method combines the remarkable radon absorption ability of the polycarbonate material of which the disks are made with its alpha track-etch properties. First proposed for radon indoors, the method can also be used for measurements in mines, water and soil-gas. Specific feature of this method is the possibility for individual a posteriori calibration of detectors and a posteriori temperature correction, as the temperature is the only known so far environmental factor that has influence on the results. This allows precise measurements of less than 20% uncertainty.

To develop this technique to its full potential a dedicated laboratory should be established. In this report we describe the performance of the novel laboratory infrastructure that has been created for that purpose in Sofia University. This includes new computerised equipment for electrochemical etching of CDs/DVDs, and a laboratory facility for calibration and experimental studies. This facility makes it possible to expose the detectors at static as well as at dynamic temperature and activity concentrations, at which they can follow controlled time functions. The temperature is measured and regulated by specially constructed programmable thermostat. Different reference activity concentrations in the exposure vessel could be made by regulating the flow-rate of the air that transfers radon/thoron activity from the sources toward the exposure vessel.

The working range of the thermostat is  $-15^{\circ} \div +50^{\circ} \text{C}$ . The range of the activity concentrations in a 50 L exposure vessel is  $0 \div 1800 \text{ kBq m}^{-3}$ . Radon and thoron are supplied by certified emanating  $^{226}\text{Ra}$  (Czech Metrological Institute, Czech Republic) and  $^{228}\text{Th}$  (Pylon Electronics Inc., Canada) sources. The concentration levels in the system are controlled through the flow-rate of air through the sources. Fig. 1 depicts the  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  concentrations vs. the air flow rate through the sources. The reference  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  concentrations are measured by calibrated radon/thoron monitors AlphaGUARD RnTn Pro (Saphymo GmbH, Germany) and RAD7 (DurrIDGE Company Inc, USA) with accuracy within  $\pm 5\%$ .

The infrastructure allows calibration of radon/thoron detectors at different temperatures that are close to those in

real exposures. In addition, it makes possible laboratory studies of the effect on the results of temperature and concentration variations that are typical for the real environment, e.g. the random variations in concentrations and temperature, diurnal variations, systematic changes similar to that possible after technological interventions etc.

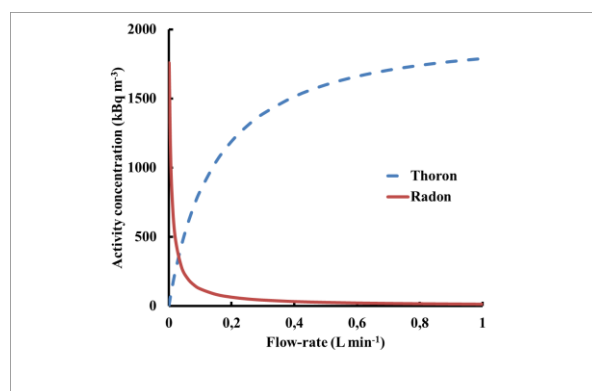


Figure 1. Reference activity concentrations at different flow-rates through the source.

The results obtained demonstrate that the capability of the infrastructure goes beyond the CD/DVD method and can potentially serve for many sorts of radon/thoron detectors. Perspectives of the use of this infrastructure encompass "traditional" calibration modes as well as research on crucial problems in environmental radioactivity like retrospective radon and thoron measurements, effect of technological interventions on indoor radon, radon in water and soil-gas etc.

## Acknowledgments

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# Monte Carlo simulation of background of HPGe detectors in deep underground installations

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An increasing number of experiments have been devoted to the detection of very rare events, e.g. in double beta-decay, dark matter and neutrino experiments, as well as in environmental physics (Arnold et al., 2005; Martineau et al., 2004; Povinec et al., 2008). Their common feature is a utilization of high sensitive low-level counting spectrometers, operating very often underground. If a detector was constructed from selected radioactivity free materials, the dominating background component in a surface or shallow underground laboratory would be cosmic rays. The flux of soft component cosmic-ray particles (electrons, positrons, gamma-rays) can be considerably decreased due to electromagnetic showers in materials with high atomic number, e.g. lead, iron, copper, etc. (Povinec et al., 2008). On the other hand, the flux of hard component particles (muons) can be decreased only by installation of detectors deep underground, or by using an antic cosmic shielding. For optimization of the background of a counting system it is useful to use a Monte Carlo simulation, so the background characteristics can be estimated in advance (Breier and Povinec, 2010)

The aim of our work has been a developing of a computing code that would allow an evaluation of background components of low-level HPGe  $\gamma$ -spectrometers optionally equipped with antic cosmic shields. The simulation code is based on the CERN's GEANT 4 package. In this work we are discussing simulations of  $\gamma$ -detectors, which have been operating in the Gran Sasso underground laboratory (3000 m w. e.).

The Monte Carlo code, which is presented in this work is consisting of three basic parts:

- i) Muon generator,
- ii) Transport of muons through a standard rock,

iii) Interaction of muons with the HPGe detector.

For verification of Monte Carlo simulations we used comparisons with experimentally measured gamma-spectra. Simulated spectra predict a lower background than experimental ones what can be due to a dominating component of background induced by radioactive impurities in the material of detectors. This component has not been included yet in our model.

## Acknowledgements

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## STUDY OF AIR BASIN NATURE AND RADIOACTIVE CONTAMINATION LEVELS AT THE SEMIPALATINSK TEST SITE (STS) AND THE ADJACENT TERRITORIES.

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Nuclear weapon tests at the Semipalatinsk Test Site resulted in extensive contamination of the environment with long-lived  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$  radionuclides. Air environment is one of ecosystem major components. Since radioactive dust raised into the environment in terms of agricultural activity is suspended, its radioactive particles may spread beyond the borders of test sites which can pose danger for the population and animals when grazing cattle.

Formerly, STS air basin long-term monitoring was not conducted, only one-time works at «Experimental Field» site and 2-year air environment in Kurchatov city. Findings were not sufficient to assess the current state of STS air basin and the adjacent territories.

The purpose of works was detection of radioactive contamination levels in the STS air basin under natural conditions and when performing production works.

To determine radioactive contamination levels of the air basin there were performed works on air environment study directly in a radioactively contaminated area, at conditionally clean territories and various distances from areas of high radioactive contamination level in soil. To assess radionuclides spreading outside STS, air basin research in populated localities adjacent to STS territories (Kurchatov city, Bodene, Sarzhal and Kaynar settlements) was carried out.

Air sampling was performed by using an electromechanical filter element air sampler (filter). At air monitoring sites, in populated localities and mineral deposits (Karadzhai, Karazhyra) a stationary air sampler was set up and the average volume of air pumped through the filter was  $n \cdot 10^4 \text{ m}^3$ . In the research areas with no permanent power supply a portable air sampler was set up and the average volume of air pumped through the filter was  $n \cdot 10^2 - n \cdot 10^3 \text{ m}^3$ . Filters were examined for presence of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$  and  $^{90}\text{Sr}$  artificial radionuclides.

In each research areas volumetric  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  radionuclides activity in the air environment did not exceed permissible volumetric activity for a population category ( $\text{PVA}_{\text{POP}}$ ). The most obvious indicator of air contamination is presence of  $^{239+240}\text{Pu}$  radionuclide. Under natural conditions at a radioactively contaminated territory  $^{239+240}\text{Pu}$  concentration does not exceed  $100 \text{ } \mu\text{Bq}/\text{m}^3$ . When having an anthropogenic impact on soil cover (soil harrowing) in terms of radioactive contamination  $^{239+240}\text{Pu}$  concentration reaches  $30000 \text{ } \mu\text{Bq}/\text{m}^3$ , which exceeds  $\text{PVA}_{\text{POP}}$  more than 10 times and is at  $\text{PVA}_{\text{PERS}}$  level. At a distance of 300 m from the area of radioactively contaminated soil cover  $^{239+240}\text{Pu}$  average concentration in the air environment does not exceed  $20 \text{ } \mu\text{Bq}/\text{m}^3$  for the whole research period.

In populated localities and mineral deposits  $^{239+240}\text{Pu}$  concentration varies from 0.4 up to  $2 \text{ } \mu\text{Bq}/\text{m}^3$  which is by a factor of 1000-10000 (3-4 orders) below  $\text{PVA}_{\text{POP}}$  level, and poses no danger to population or personnel who are performing agricultural and production works at the above territories.

The research conducted showed that agricultural works being performed and unrelated to anthropogenic impact on soil cover, cannot have a negative effect on the air environment. When having an impact on soil cover (soil harrowing) transuranic radionuclides concentration in the air environment is at  $\text{PVA}_{\text{POP}}$  level, such works must be performed using individual protective equipment and constant radiation control.

# Sequential separation of Pu isotopes and $^{90}\text{Sr}$ in seafood using ion-exchange/extraction chromatography

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**Keywords:** food,  $^{90}\text{Sr}$ ,  $^{239,240}\text{Pu}$ , ion exchange, extraction chromatography

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A rapid and straightforward method has been developed to determine Pu isotopes and  $^{90}\text{Sr}$  in seafood. The study was commissioned by Ministry of Food and Drug Safety of Korea, formerly known as Ministry of Food and Drug Safety (MFDA), after the Fukushima nuclear disaster in March 2011 in order to cope with the large number of samples in short period of time. This study is to establish a rapid and simple radioanalytical method to determine the Pu and Sr isotopes in the seafood stuffs and to evaluate an impact of the Fukushima Daiichi NPP accident on the Korean seafood stuffs.

The new procedure combined two separate procedures for Pu isotopes and  $^{90}\text{Sr}$  into one simple method. Anion resin column was placed on the top and Eichrom's Sr-resin column was placed directly under the anion resin column. A sample was loaded in 8M nitric acid followed by couple of acid wash steps and the Sr-resin column was removed for  $^{90}\text{Sr}$  extraction. Pu isotopes were eluted from the anion resin column using  $\text{NH}_4\text{I}/\text{HCl}$  reagent and  $^{90}\text{Sr}$  was eluted using Milli-Q water.

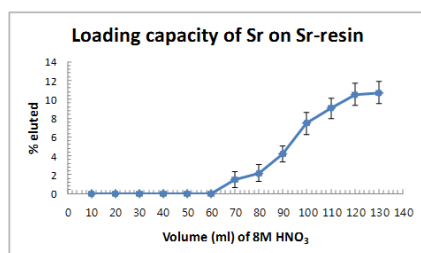


Figure 1 Behaviour of Sr in 4cm Sr-resin column

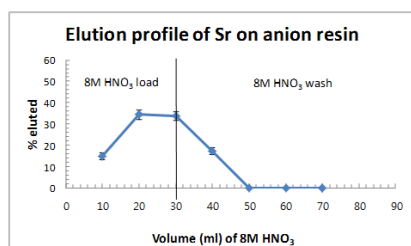


Figure 2 Behaviour of Sr in 5cm anion resin column

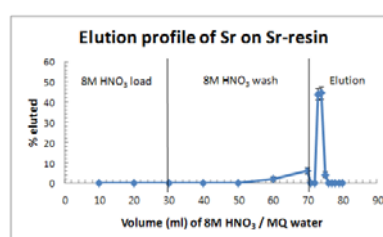


Figure 3 Optimised separation method for Sr from Sr-resin column

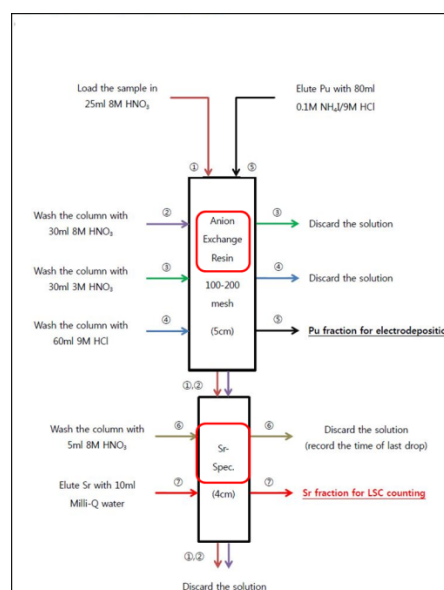


Figure 4 Flow chart of sequential separation of Pu and Sr by anion exchange resin and Sr resin

Reproducibility test as well as the test using tracer solutions showed that the method would be very reliable for seafood samples. Recoveries turned out to be high, > 90% for Pu isotopes and > 80% for  $^{90}\text{Sr}$ , respectively.

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# First radioactivity monitoring of soil in Serbia

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**Keywords:** soil radioactivity monitoring, gamma spectrometry, effective dose, strontium

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The results of first comprehensive soil radioactivity monitoring study in Serbia are presented and analysed in this paper. The study involved two methods, gamma spectrometry measurement of 72 soil samples (agricultural and non-agricultural) from 24 different locations and determination of <sup>90</sup>Sr in 8 samples of non-agricultural soil. The monitoring was performed during autumn 2014 by two accredited and authorized institutions for these types of measurements: Radiation and Environmental Protection Department, Institute Vinča from Belgrade and Laboratory for Radioactivity and Dose Measurements, Faculty of Sciences, from Novi Sad. Monitoring was supported by the Serbian Radiation Protection and Nuclear Safety Agency.

Soil samples were taken from two different depths: agricultural soil from surface layer (0 – 20 cm) and non-agricultural soil from two depth layers (0-5 cm and 5-15 cm).

The samples were dried at 105°C, milled, homogenized and sealed in two geometries: Marinelli beakers of 0.5 l and cylindrical vessels (62 mm x 67 mm) according to method of measurement [1]. In order to establish radioactive equilibrium of radon and their progenies, samples were prepared and sealed for 40 days, prior to analysis. Gamma spectrometry measurements were performed by low-level HPGe detectors with high resolution in different shields.

Method for determination of the <sup>90</sup>Sr means the radiochemical separation of yttrium from the sample, so the activity of <sup>90</sup>Sr is determined on the basis of measurements of <sup>90</sup>Y after the establishment of radioactive equilibrium <sup>90</sup>Y/<sup>90</sup>Sr. The measurements of strontium were performed on a gas proportional counter.

The minimum values of the activity concentration of natural radionuclides were detected in agricultural soil on the site Zlatibor, while on the other locations variations in the values of activity concentrations of natural radionuclides were not noticed. The ratio of <sup>235</sup>U / <sup>238</sup>U in the range of 0.041 to 0.057 corresponds to the natural origin of uranium.

Distribution of measured activity concentrations of natural and artificial radionuclides are given and discussed in the paper.

The minimum value of <sup>90</sup>Sr activity concentration was detected in a sample of soil at the site of Niš, while the maximum concentration was detected in soil samples at the site of Kladovo.

Estimated values (minimum and maximum) of the absorbed dose of gamma radiation (D) and annual effective dose DE (minimum and maximum) of external exposure to gamma radiation from agricultural and non-agricultural soil are listed in Table 1.

Table 1. Absorbed and effective doses due to different depths and types of soil.

Non-agricultural	D [nGy/h]	DE [mSv/a]
0 – 5 cm	21,3 – 83,9	0,026 – 0,103
5 – 15 cm	20,3 – 78,6	0,025 – 0,096
Agricultural 0-20 cm	39,2 – 83,1	0,037 – 0,102

The maximum activity concentration of artificial radionuclide <sup>137</sup>Cs was detected in the surface layer of agricultural soil on the site Prijepolje (7), and the minimum values at locations Smederevo (10) and Šabac (12) (Figure 1). <sup>137</sup>Cs depth profiles for agricultural and non-agricultural soil were compared and discussed in the paper.

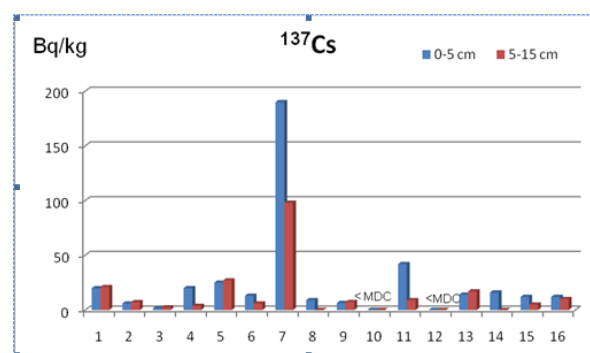


Figure 1. Measured activity concentrations of <sup>137</sup>Cs in non-agricultural soil samples from different depths (0-5 cm and 5-15 cm)

## Acknowledgments

The investigation was partially supported by the Ministry of Science and Environmental Protection of Serbia under the following Projects 43009 and 43002.

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- [1] IAEA Technical Reports Series No.295 – Measurement of Radionuclides in Food and the Environment-Section 5-Collection and Preparation of Samples

# Multi-criteria optimization tool for the decontamination of urban areas after a nuclear accident

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**Keywords:** multi-criteria process, nuclear accident, urban area, decontamination.

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This work describes the development of the multi-criteria program designed to rank decontamination procedures considering technical and radiological protection aspects to support decisions on remediation of urban areas contaminated with Cs-137.

The first stage was the development of a database on decontamination procedures, including the main characteristics to be used as part of the criteria for a decision. This work was done, basically, by literature review. The database includes 25 decontamination procedures that may be applied to urban environments. The procedures were classified according to their main characteristics. The description of each procedure includes [Silva et al., 2009]:

- (a) General aspects: description of the procedure, type of surface to be treated, radionuclides to which the procedure has been developed, scale of application;
- (b) Radiological protection: exposure pathways, efficiency on removing contamination, doses to workers and technical restrictions to its application;
- (c) Infrastructure needed, including equipments, materials, specialized workforce and safety requirements;
- (d) wastes: Qualitative and quantitative aspects.

For the multi-criteria decision tool, two types of criteria need to be defined. The subjective criteria were derived from questionnaires distributed to specialists on accident management [De Luca et al., 2013]. The technical criteria are being developed by two different procedures. Those that are independent of the scenario are included in the model as default values. These include the needs on infrastructure, type of wastes generated and the need of trained people. Those that depend on the scenario are assessed through simulations of accidents and the consequences of applying each countermeasure described in the database [Silva et al., 2013b] to pre-defined scenarios. The scenarios for typical tropical residential urban environments were selected based on images from Google Earth® for the 50 km area surrounding the Brazilian Nuclear Power Station. For the simulation of public exposure on these scenarios, occupancy rates and number of people per unit are raised. For occupational doses, the time and number of workers needed to accomplish each task are considered [Silva et al., 2013a].

The simulation of accidents to assess environmental concentration and averted doses to members of the public due to the application of countermeasures were performed using a software based on PARATI model [Rochedo et al., 1998].

Occupational doses and amounts and concentrations of wastes are performed by specific models developed and included in the software package [Silva et al, 2015].

Finally, the procedures are classified according to the different criteria and feed to the multi-criteria tool software model. For each procedure, each subjective criterion value is multiplied by the average value of the associated technical criteria and summed. The output is a rank list of procedures optimized according to the established criteria. The overall structure of the model is shown in Figure 1.

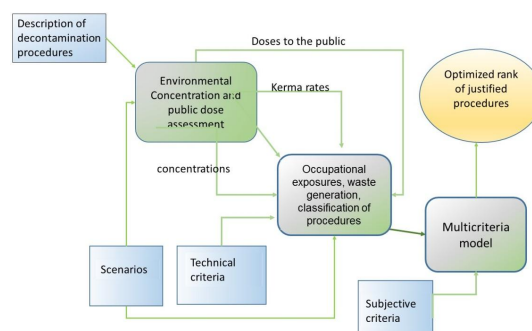


Figure 1. Schematic presentation of the model modules

The development of numerical values aiming to perform optimization procedures shall continue to include rural areas and surface water environments. It must be emphasized, however, that there are criteria that cannot be established in advance as they shall depend on the actual situation. Also, direct costs and public concern aspects are not included in this work as these aspects shall be covered by other institutions participating of a decision-making process.

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## Effect of tritium on bio- and photoluminescence of blue fluorescent protein

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*Keywords:  $\beta$ -radiation, tritium, blue fluorescent protein, fluorescence spectra*

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Tritium, a beta-emitting isotope of hydrogen, is one of the most widespread products of radioactive decay of industrial radioisotopes. Due to low energy of its radiation, tritium produces a specific action on living organisms.

To study effects of tritium on organisms, we chose photoprotein obelin, a representative of a group of blue fluorescent proteins. Obelin is a perspective bioluminescent and fluorescent biomarker for biomedical investigations. As a biomarker, it has several advantages, such as stability, non-toxicity and high fluorescence quantum yield.

Addition of  $\text{Ca}^{2+}$  to the photoprotein obelin triggers a bioluminescent reaction with light emission. The product of the bioluminescent reaction – enzyme-bound coelenteramide – is a blue fluorescent protein called ‘discharged’ obelin. Bioluminescent spectra of obelin and photo-luminescent spectra of discharged obelin include several components (emitters), corresponding to different forms of coelenteramide (Belogurova *et al.*, 2008). Contributions of these components determine a color of the bio- and photoluminescent spectra of obelin. The contributions to the photoluminescence spectra can vary under influence of various physical and chemical factors – high-temperature exposure variation of excitation wavelength, concentrations of calcium ions or biochemical agents (Belogurova *et al.*, 2010; Alieva *et al.*, 2013, 2014).

In this work we studied effects of tritium on spectral characteristics of bio- and photoluminescence of obelin.

Reagent: lyophilized preparation of  $\text{Ca}^{2+}$ -regulated photoprotein obelin and Tris-HCl buffer, pH 7.0. Tritiated water (HTO), 200 MBq/L, was used as a medium for the bioluminescent reaction and as a source of ionizing radiation. Bioluminescent reaction was initiated by  $\text{Ca}^{2+}$ . The product of the bioluminescent reaction, discharged obelin, was incubated in HTO for 192 h at 5°C. Bioluminescence spectra and photoluminescent spectra of discharged obelin were registered. Photoluminescent spectra were measured every 24 h after the bioluminescent reaction was ceased. The 280 and 350 nm excitation and 500 nm registration wavelengths were used. The spectra were compared to those of a control sample in nonradioactive solution. All the photoluminescent spectra were deconvolved into components using Gauss-based distribution and method of second derivative.

It was found that the bioluminescent spectra of obelin were not sensitive to the presence of HTO.

Photoluminescent spectra of the discharged obelin at 280 nm photoexcitation included several peaks – 350, 410 and 660 nm. It was found that HTO suppressed the obelin photoluminescence intensity. Additionally, 500-nm maximum in the green spectral region appeared and increased its intensity with the time of exposure, while the “violet” 410-nm peak was suppressed. As a result, the spectral composition of the photoluminescent spectra of obelin (lyophilized preparation) measured in HTO was closer to this of fresh obelin preparation.

The decrease of photoluminescent obelin intensity was explained with denaturation ability of HTO, while “greening” of the obelin photoluminescence was concerned with higher efficiency of charge transfer process in obelin chromophore in the radioactive solution.

Thus, the exposure of obelin to HTO, 200 MBq/L, did not change obelin bioluminescence intensity and color. However, it decreased intensity of photoluminescence of discharged obelin (blue fluorescent protein, the product of the bioluminescent reaction) and changed its color: the contribution of blue spectral component decreased, while the contribution of green component increased.

The study forms a basis for development of fluorescent biomarker to monitor biological toxicity of radioactive media.

### Acknowledgments

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# Estimation of $^{131}\text{I}$ deposition from $^{129}\text{I}$ analysis in surface soils released from the Fukushima Daiichi Nuclear Power Plant accident

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**Keywords:**  $^{131}\text{I}$ ,  $^{129}\text{I}$ , FDNPP accident, AMS.  
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Radioiodine is one of the most important radionuclides released from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. The total amount of  $^{131}\text{I}$  discharged in environment was estimated to be approximately  $2.0 \times 10^{17}$  Bq (Kobayashi et al., 2013).  $^{131}\text{I}$  has the tendency to accumulate in the human thyroid gland. However,  $^{131}\text{I}$  (half-life: 8.02 d) has a short half life time, it is therefore difficult to estimate the radiation dosimetry for  $^{131}\text{I}$  at this time. It is necessary to obtain information regarding the deposition of  $^{131}\text{I}$  during the initial stage in different locations after the FDNPP accident at Fukushima.

There are a few data related to  $^{131}\text{I}$  deposition on the ground released from the FDNPP accident (Kinoshita et al, 2011; MEXT, 2011). On the other hand,  $^{129}\text{I}$  (half-life:  $1.57 \times 10^7$  y) was also released from the FDNPP accident.  $^{129}\text{I}$  and  $^{131}\text{I}$  are same element and transport identically in environment. It is expected to estimate  $^{131}\text{I}$  deposition from  $^{129}\text{I}$  with the long half-life time in order to improve the precision of  $^{131}\text{I}$  estimation. The atomic ratios of  $^{129}\text{I}/^{131}\text{I}$  were previously reported in Miyake et al. (2012) as  $31.6 \pm 8.9$  and Muramatsu et al. (2015) as 20.8 for the FDNPP accident.

We measured  $^{131}\text{I}$  concentrations in surface soils at 108 sites around the FDNPP immediately after the accident by gamma-ray spectrometry (Kinoshita et al., 2011). In this study, we used same surface soils.  $^{129}\text{I}/^{127}\text{I}$  ratios were measured by accelerator mass spectrometry (AMS) at the MALT, the University of Tokyo (Matsuzaki et al., 2007). Stable iodine of  $^{127}\text{I}$  was determined by the inductively coupled plasma mass spectrometry (ICP-MS). We got a result that the average  $^{129}\text{I}$  concentration was  $(2.7 \pm 1.4) \times 10^8$  atoms/g prior to the FDNPP accident as  $^{129}\text{I}$  background at Fukushima. After the accident, average isotopic ratio of  $^{129}\text{I}/^{131}\text{I}$  is estimated to  $26.0 \pm 5.7$  as at March 11, 2011. Figure 1 shows the relationship between the concentrations (atoms/g) of  $^{129}\text{I}$  and  $^{131}\text{I}$  in surface soils at Fukushima. There is a good correlation between the concentrations of  $^{129}\text{I}$  and  $^{131}\text{I}$  ( $R^2 = 0.98$ ). We found the difference of  $^{129}\text{I}/^{131}\text{I}$  ratios depending on the locations. It is thought to be due to reflect differences related to emission sources from the FDNPP. The result of calculations about  $^{129}\text{I}/^{131}\text{I}$  ratios made by the ORIGEN2 code are 31.4 for the Unit 1 reactor, 21.9 for the Unit 2 reactor and 20.8 for the Unit 3 reactor (Nishihara et al., 2012).

In this presentation, we report the distribution of  $^{129}\text{I}$  in terrestrial environment at Fukushima and  $^{129}\text{I}/^{131}\text{I}$

ratios by region. We also make a comparative review of previous works and the ORIGEN2 code calculations with our results. In addition, we try to reconstruct  $^{131}\text{I}$  deposition density at Fukushima using the analytical results of  $^{129}\text{I}/^{131}\text{I}$  ratios in surface soils.

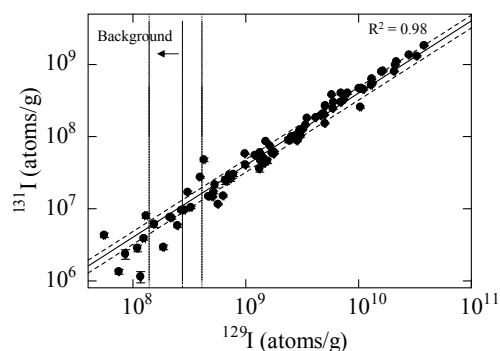


Figure 1. Relationship between the concentrations (atoms/g) of  $^{129}\text{I}$  and  $^{131}\text{I}$  in surface soils at Fukushima.

## Acknowledgments

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# Airborne monitoring in the course of nuclear accident scenarios and in post accidental environmental conditions

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**Keywords:** airborne monitoring, NPP accidents, radionuclides release into environment, doses from radioactive cloud.  
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Airborne monitoring and airborne gamma spectrometry are well known as the powerful tools for the prompt monitoring and mapping of the large areas to explore contamination caused by the nuclear facilities operation as well as accidents with radionuclides leakages. In the specific scenarios of the NPP severe accidents the airborne monitoring (including drones) proved to be (Fukushima accident, Chernobyl) useful means of monitoring the radiation situation in the NPP area and close vicinity during the process of accident and remediation measures. Aerial vehicles were used also for measures and interventions geared towards reduction, mitigation and remediation of the accident impacts.

Aerial vehicles can be divided into category of the piloted and unmanned ones – drones. In the first category it is necessary to evaluate the doses received by the crew and monitoring/intervene personnel from cloud of air contaminated by the radionuclides accidental leakage to ensure adequate radiation protection. From the radiation protection viewpoint also the evaluation of the shielding factors of the considered aerial vehicles is desirable. For the second category the assessment of dose rates in the operational space in case of severe accidents can be important from point of view the radiation resistance of on-board equipment.

Assessment of dose rates inside and outside of the cloud of contaminated air for different scenarios of the NNP accidents, leakages and spreading conditions was done using MC simulation (MCNPX code). Technique similar to calculations (Kluson et al., 2010) was used. Example of calculated energy dependence of cumulative distribution of the photon flux in the centre of homogeneous spherical cloud is in the Figure 1.

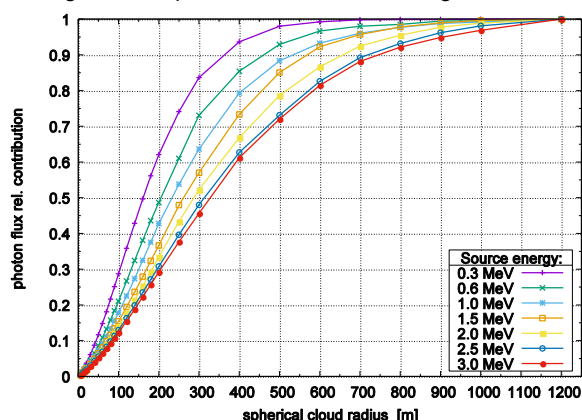


Figure 1. Calculated contributions of the cloud layers to the photon flux in the cloud centre.

To assess the shielding factors of the helicopters used or considered for the airborne monitoring the simplified models of the fuselage and cockpit were described and model calculations using MCNPX code were done. Example of the helicopter fuselage simplified model is in the Figure 2.

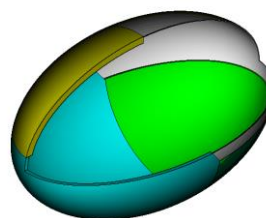


Figure 2. Simplified model of the helicopter fuselage.

Resulting very low shielding capabilities of the considered helicopters prove the expected conclusion that shielding by air plane has marginal meaning for radiation protection taking into account uncertainties in the knowledge of the cloud configuration.

For the post accidental an environmental monitoring the airborne gamma spectrometry can be used. The unfolding technique used calculated detector response matrix enables to determine the contaminant concentrations and corresponding dose rates on the ground (Kluson, 2010 and Kluson et al., 2015). The method and results of the calculations of response matrix and dependence of the space resolution on the altitude of flight are presented and discussed and uncertainties of the airborne monitoring analysed.

## Acknowledgments

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# Sr-90 measurement in Japanese soil samples after the Fukushima nuclear accident

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**Keywords:** Sr-90, Cs-137, LSC, Fukushima, Soil.

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The cooling system of the nuclear reactors were damaged by a devastating tsunami hitting the Fukushima Daiichi Nuclear Power Plant (FDNPP), Japan, which resulted in a nuclear major accident (Level 7 of the International Nuclear and Radiological Event Scale) with international radioactive contamination. Among the released volatile fission products (I, Te, Cs isotopes), intermediate volatile elements such as Sr and Ru isotopes also were emitted.

Sr-90 is a biologically hazardous radionuclide considering its similar metabolism in human tissues to the calcium. Bone cells can uptake strontium from the blood and recreate the bone tissue wherein the decaying Sr-90 and its decay product Y-90 can cause internal radiation exposure. Therefore, the environmental monitoring to reveal the contaminated areas in Japan is essential.

In this study, soil samples with relatively high Cs-137 were collected from the vicinity of the damaged Fukushima Daiichi Nuclear Power Plant. The soil samples were chemically open digested using an acid mixture of HNO<sub>3</sub>, HF and HClO<sub>4</sub>. The strontium separation was carried out using extraction chromatography resin (Sr resin) and Sr-90 was detected with a liquid scintillation counter system (TriCarb-3100).

The level of activity concentrations of Sr-90 in the analysed soil samples was no more than tens of Bq kg<sup>-1</sup> showing good agreement with the results of recently

published surveys (Kavasi, 2015, Mishra, 2014). It has been reported in Japan before the FDNPP about 6 Bq m<sup>-2</sup> in soil might be originated from atmospheric nuclear weapon tests. In this study, we did not notice any significant Sr-90 contamination from the limited samples and internal radiation dose caused by Sr-90 uptake could be negligible.

## Acknowledgments

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# Atmospheric deposition characteristics of $^7\text{Be}$ at Málaga (South Spain)

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Keywords:  $^7\text{Be}$ ; wet deposition; Trajectory analysis  
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The radioactive, water soluble and aerosol-borne substances such as  $^7\text{Be}$  act as natural radionuclide tracer of aerosols originating over a range of altitudes in the atmosphere and are considered as useful tools for studies such as atmospheric deposition, cloud formation and global transport processes. This cosmogenic radionuclide is produced in the stratosphere and troposphere, and enhanced mixing between the stratosphere and troposphere with the mid-latitude tropopause folding during the spring has been shown to increase its presence in the troposphere. Intense storms also can transport  $^7\text{Be}$  from the upper to lower troposphere, increasing the availability of  $^7\text{Be}$  that can then be scavenged through precipitation.

Bulk atmospheric deposition of gamma radionuclides is continuously measured on a monthly basis at the University of Málaga (4° 28' 80" W; 36° 43' 40" N) which is one of the environmental radioactivity monitoring network stations operate by the Spanish Nuclear Security Council (CSN) in South Spain (Dueñas et al., 2005). In this study, the results of an experimental research project carried out to add new details to the patterns of depositional fluxes of this radionuclide in this coastal Mediterranean site are reported. Knowledge of the behavior of  $^7\text{Be}$  in the atmosphere would yield insight on the behavior of other similar chemical species in the atmosphere. Therefore, dry and wet-deposition samples were collected at this station using an automatic sampler and additionally, concentrations of  $^7\text{Be}$  in air in periods of wet sampling were also recorded using a high-volume air sampler. Deposition fluxes and atmospheric concentrations of  $^7\text{Be}$  were observed from October 2012 to March 2015 (Fig. 1 and Fig. 2). A total of 26 wet only samples were collected during the study period and bulk weekly samples were also obtained for comparison. Precipitation in Málaga is highest from November to January while almost non-existent in July and August. Some of the samples were collected from a single event of rain and when there was less amount of rain in an event or events, then the samples of two or more events in a week were combined to form a larger sample. All the radiometric measurements were performed by low-level gamma spectrometry with a coaxial-type germanium detector and it was calibrated using certified reference gamma ray cocktail.

A significant positive correlation between precipitation and the atmospheric fluxes of  $^7\text{Be}$  was found and are also similar to previous studies in the region. Dry fallout of  $^7\text{Be}$  was found to be very small compared with wet fallout. Intense rainfall may serve to deplete partially the upper troposphere of  $^7\text{Be}$  such that later events occurring close in time display lower fluxes despite

similar rainfall amounts. Air flow patterns arriving at Málaga were identified by back-trajectory analysis using the HYSPLIT model (Draxler and Rolph, 2013). 96-hour kinematic 3D back-trajectories starting at 0, 6, 12 and 18 UTC each day during the study period at three heights (500, 1500 and 3000 m a.s.l.) were computed. The origin and the history of the air masses determine to a great deal the concentration of  $^7\text{Be}$ -bearing aerosols, and the precipitation at the study area and along the route to the study area.

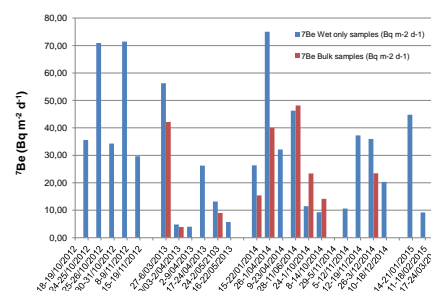


Figure 1. Deposition fluxes of  $^7\text{Be}$  in wet only and bulk samples.

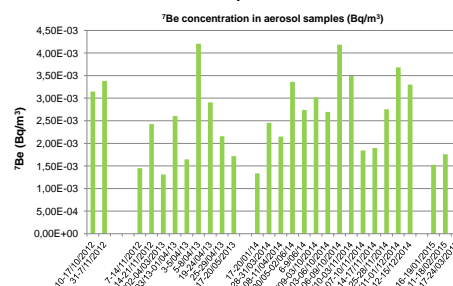


Figure 2. Concentration of  $^7\text{Be}$  in aerosol samples.

## Acknowledgments

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# Remote controlled aerosol sampler with gamma spectrometric module – design, functionality and initial working experience

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Keywords: atmosphere, aerosol sampling, artificial radionuclides, gamma spectrometry

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## INTRODUCTION

Early and sensitive detection of the increased levels of artificial radionuclides in the atmosphere by a high volume aerosol sampling device (HVS) is solved in the MOSTAR research project.

A remotely controlled HVS device with an online gamma spectrometric unit above the aerosol filter was designed and prototyped. Research objectives were focused mainly on the optimal design with respect to the spectrometer choice, measurement geometry and spectra evaluation algorithm to provide the lowest detection limits possible.

This work presents the design, functionality and initial working experience with the device prototype. Some of the preliminary results regarding its possible use for atmospheric radon dynamics research are also discussed.

## MATERIALS AND METHODS

Four types of spectrometers (HPGe, NaI(Tl), LaBr, CdZnTe) were placed above the HVS filter during aerosol sampling and their energetic resolution, detection efficiency and sensitivity to the radionuclides expected after a NPP accident such as <sup>131</sup>I, <sup>132</sup>I, <sup>132</sup>Te, <sup>134</sup>Cs and <sup>137</sup>Cs was compared.

Quantification of the radon effect on the detection limits was assessed by a series of sequential measurements under different radon volume activity (RVA) conditions in the sampled air. Based on these measurements, the mathematical model which allows predicting the minimum detectable activity (MDA) of the radionuclides of interest was constructed:

$$MDA_i = f(k, t) \cdot \sqrt{RVA} + MDA_{BG_i}$$

$MDA_i$  stands for the MDA of the  $i$ th radionuclide,  $f(k, t)$  is the scaling function dependent on the <sup>222</sup>Rn/<sup>220</sup>Rn activity ratio  $k$  and sampling duration  $t$ ,  $MDA_{BG_i}$  is the MDA background level for the  $i$ th radionuclide.

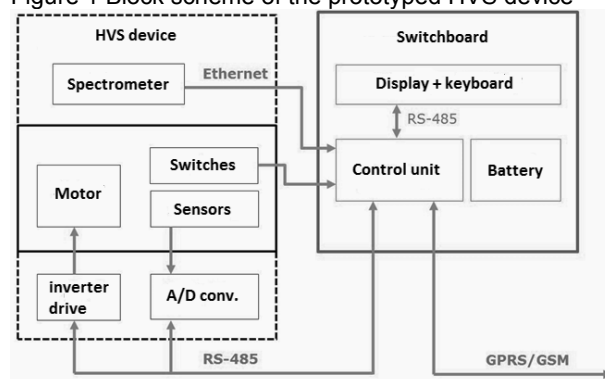
## RESULTS AND CONCLUSIONS

Based on preliminary experiments, a remotely controlled HVS device was prototyped. The device communicates (via GPRS/GSM/LAN) with the central server accessible through the web based GUI, where technological and spectrometric data are stored and available to the users.

User can easily set the device flow rate in the range of 15 – 150 m<sup>3</sup>/h, parameters of spectra acquisition and reference level for the alarm initiation.

Device status and measured values are sent via SMS or email to the operators.

Figure 1 Block scheme of the prototyped HVS device



Due to significant fluctuations of the RVA in the atmosphere, it was necessary to design and implement robust heuristic algorithm which minimizes the number of false positive alarms and is sensitive enough at the same time.

Table 1 Detection limits for a typical 1 h measurement, NaI(Tl) spectrometer and selected radionuclides

RN	Half-life	Energy [keV]	Intensity [%]	MDA [Bq/m <sup>3</sup> ]
<sup>131</sup> I	8.0 days	364	81.2	0.6
<sup>132</sup> I	2.3 hours	668	98.7	0.5
<sup>132</sup> Te	3.2 days	228	88.1	0.6
<sup>134</sup> Cs	2.1 years	605	97.6	0.5
<sup>137</sup> Cs	30.1 years	662	85.0	0.7

Comment: MDA values are site specific since they depend on the RVA.

Device modular concept enables the end user to easily change the spectrometer type. The NaI(Tl) detector was chosen for the prototyped device as a compromise between the price, sensitivity and outdoor use endurance. At higher costs, a better alternative would be the LaBr spectrometer which is superior to the NaI(Tl) in energy resolution which is an important factor as lower detection limits would be achieved.

## Acknowledgments

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# $^{210}\text{Pb}$ in size-segregated aerosol samples in Northern Finland

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**Keywords:** impactor, lead-210, aerosols, Finland.

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Lead-210 ( $^{210}\text{Pb}$ ) is formed in the atmosphere from the radioactive noble gas  $^{222}\text{Rn}$  emanating from the Earth's crust. 99 % of the airborne  $^{222}\text{Rn}$  originates from land and only 1 % from the sea and consequently high  $^{210}\text{Pb}$  concentrations are found in continental air masses (Baskaran, 2011). Owing to the long half-life (22.3 y) of  $^{210}\text{Pb}$ , its removal from the atmosphere is governed by the wet and dry deposition processes affecting the aerosol particles carrying it rather than radioactive decay. Lead-210 decays to polonium-210 ( $^{210}\text{Po}$ ,  $t_{1/2} = 138.4$  d). Being an alpha emitter  $^{210}\text{Po}$  is highly radiotoxic, especially when inhaled. The size distribution of aerosol particles carrying  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  is crucial for two reasons, atmospheric behavior and radiation protection. The efficiency of atmospheric removal processes depend on the size of aerosol particles carrying  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ . In some cases the aerosol particle size also affects its atmospheric transport properties. From the radiation protection point of view the size distribution of aerosol particles carrying  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  determines the ability of these particles to penetrate into the human respiratory system. The objective of this work was to determine the aerodynamic size distribution of aerosol particles carrying  $^{210}\text{Pb}$  in the subarctic atmosphere of northern Finland and to study the related atmospheric effects on their concentration.

Size-segregated aerosol samples were collected in open air 2 m above the ground at the Finnish Meteorological Institute's Arctic Research Centre at Sodankylä, Finland. The site is situated 100 km north of the Arctic Circle at 67.368°N, 26.633°E, and 179 m above mean sea level. The aerodynamic size distribution of aerosol particles carrying  $^{210}\text{Pb}$  was obtained by using a high-volume 6-stage cascade impactor with efficient cutoff diameters of 0.39, 0.69, 1.3, 2.1, 4.2 and 10.2  $\mu\text{m}$ . The length of each collection period was 48 h. Glass fiber filters were used as impaction substrates and back-up filters. The samples were analysed for  $^{210}\text{Pb}$  at the Laboratory of Radiochemistry, University of Helsinki with the classical method of spontaneous deposition of polonium onto silver disks and subsequent alpha spectrometry (Flynn, 1968).

The relative activity concentration of  $^{210}\text{Pb}$  in different particle size bins shows that most of the  $^{210}\text{Pb}$  is attached to accumulation mode aerosol particles with an

aerodynamic diameter below one micrometer (Fig. 1). In summer  $^{210}\text{Pb}$  tends to be in even smaller particles than in spring. This may be related to the longer residence time of aerosol particles in the air in spring compared to that in summer. In spring aged aerosols ("Arctic haze") are found in the atmosphere after the winter. In summer, however, the aerosols are relatively fresh and have not had yet time to grow to bigger size classes.

In an earlier work it was observed that most of the atmospheric deposition of  $^{210}\text{Pb}$  in Finland occurs via wet removal processes (Paatero et al., 2015). This is confirmed by the results of this study as most of the  $^{210}\text{Pb}$  activity is carried by accumulation mode aerosol particles which are too small to be removed by gravitation or inertial impaction but too big to be removed by diffusion.

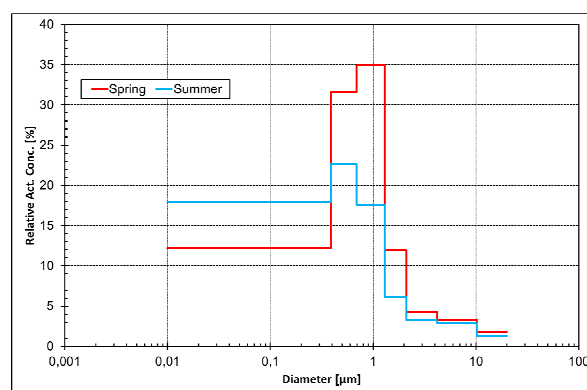


Figure 1. Average relative activity concentration of  $^{210}\text{Pb}$  (%) in different aerosol particle size classes. The lower and upper ends of the distribution are artificial.

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## Determination of Pannonian basin (Serbia) groundwater ages using tritium radioactive isotope

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**Keywords:** tritium, groundwater, age, Pannonian basin

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Tritium is a radioactive isotope of both natural and anthropogenic origin. The anthropogenic tritium originated from a series of nuclear explosions since 1952 onwards. Maximum concentration of tritium in the atmosphere was registered in 1963 up to 6000 T.U. After military nuclear tests have been stopped, concentration of tritium gradually decreased because of short half-life (12.4 years) and exchange among various reservoirs. Maximal tritium concentration in precipitation in Serbia in 2005 was about 35 TU, with the average value of 8.35 TU at location Zeleno brdo (the reference meteorological station) (Janković et al, 2012). This value was comparable to the average tritium concentrations at Zagreb (Croatia) and Ljubljana (Slovenia) (Vreča et al, 2006).

Tritium is directly incorporated into the water molecule (H<sup>3</sup>HO), so it participates in the hydrological cycle and can be detected in groundwater. The presence of tritium in groundwater indicates recharge of aquifer by modern precipitation (i.e., recharge after 1952), while age of older groundwater can be determined by using carbon isotope <sup>14</sup>C.

Modern groundwater is therefore renewable, as opposed to the older and fossil water, and in this sense it can be better utilized. On the other hand, modern groundwater is more vulnerable to contamination from the surface. It is important to know the age of groundwater for better water resources planning and management.

From the hydrogeological point of view, in the Pannonian basin (Serbia) four hydrogeological systems are identified (Aksin and Milosavljević, 1982).

First hydrogeological system is widely spread in Serbian part of the Pannonian basin, except at Mt. Fruška gora and Vršачki breg. Aquifer is composed of sands and gravels. This system is the most important for water supply of the population and irrigation.

Aquifer of the II hydrogeological system is composed of sandstone and marl.

Aquifer of the III hydrogeological system is composed of sandstone, conglomerate, limestone and breccia, while aquifer of the IV hydrogeological system is

composed of fractured and cavernous limestone and dolomite.

Tritium was determined in 29 samples of groundwater and one sample of rain water. The largest number of groundwater samples have been taken from the first hydrogeological system, where the highest tritium concentration was expected.

Water samples were electrolytically enriched with an average enrichment factor of  $23.6 \pm 2.3$  (Barešić et al, 2011). Measurements were performed by Ultra-low-level liquid scintillation counter Quantulus 1220. The scintillation cocktails contained 8 mL of enriched water sample and 12 mL of scintillator Ultima Gold LLT.

Results of samples from all hydrogeological system in the Pannonian basin can provide information about the depths of modern groundwater circulation and their spreading.

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# Radon concentration in a naturally ventilated radioactive waste interim storage facility in Greece

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**Keywords:** Radon, Radioactive Waste storage, wind, natural ventilation.

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Radioactive waste is commonly stored in interim storage facilities, awaiting for its final disposal. For this purpose dedicated surface buildings are often utilized. One of the radioisotopes often encountered in non-nuclear radiation activities is <sup>226</sup>Ra, which originates from medical, research and industrial sector and also exists in consumer products, such as lighting rods and radium illuminating dials. For <sup>226</sup>Ra sources, besides the radiation hazards connected to the isotope itself, considerations shall be also given to its daughter isotope <sup>222</sup>Rn, which is released in gaseous form and may contribute considerably to the dose of the facility staff through inhalation.

In Greece, radioactive waste is stored in the radioactive waste interim storage facility of the National Centre for Scientific Research "Demokritos". The accurate determination of the <sup>226</sup>Ra inventory stored in the facility is a laborious work due to the lack of pertinent information for the historical waste. According to preliminary rough estimations, based on the number and type of objects-sources, the total activity of <sup>226</sup>Ra stored in the facility may reach a few hundred MBq.

There are no mechanical means for ventilation in the building. Therefore, the factors dominating the radon concentration and its time fluctuations are considered to be the radon release rate from the radium sources and the building natural ventilation rate. Radon concentration in the facility is measured as a function of time, from 28 March 2014 to 01 April 2014, using a portable AlphaGUARD monitor. Measured data for the external wind are also available for the above period from NCSR "Demokritos" meteorological stations. The measurements show dependence between the radon concentration fluctuations and the variation of the external wind. Attempt is made to reproduce this dependency theoretically, using simple modelling approaches, based on the assumption of a single envelope building, with uniform radon concentration. In this case, the radon conservation equation in the facility can be written as follows:

$$\frac{dC}{dt} + \left( \frac{Q}{V} + \lambda \right) C = \frac{S}{V} \quad (1),$$

where,  $C$  (Bq/m<sup>3</sup>) is radon concentration,  $Q$ ,  $V$  and  $\lambda$  are the ventilation rate (m<sup>3</sup>/s), building volume and radon decay constant, respectively, and  $S$  (Bq/s) the radon gas release rate from stored <sup>226</sup>Ra decay. Airflow through the building penetrations is assumed to occur by natural cross-ventilation driven by wind (e.g. Freire et al. 2013).

In Figure 1, the time variation of radon concentration in the facility is shown, together with the measured wind velocity. The results from the analytical and numerical solution of Eq. (1) are also shown. For the numerical solution the time varying measured wind data are used, whereas, for the analytical solution a constant wind velocity is assumed, equal to the average value of the measurements. The release rate of radon gas ( $S$ ) was adjusted, after some trials, in order to fit the measured data. The value of the radon release rate was found to be more than one order of magnitude lower compared to the release rate that corresponds to the estimated <sup>226</sup>Ra stored activity, indicating strong radon retention in the drums. Once, however, the release rate is adjusted, the numerical model performs surprisingly good for the longer part of the examined period, as it is shown from the comparison of the calculated results with the measured data. Modeling will be further tested against additional measurements for different time periods.

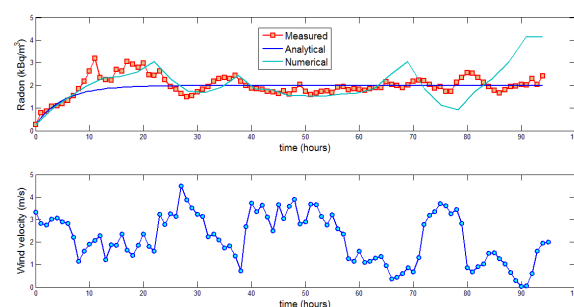


Figure 1. Calculated and measured Radon concentration in the radioactive waste storage facility, and measured wind velocity, as a function of time, from 28 March 2014 to 01 April 2014.

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# $\beta/\gamma$ -tracer of dangerous natural phenomena and technogenic accidents

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**Keywords:**  $\beta/\gamma$ -tracer, natural radioactivity, technogenic accident.  
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A new radiation  $\beta/\gamma$ -tracer was suggested for revealing of dangerous natural phenomena and technogenic accident.  $\beta/\gamma$ -tracer determines a ratio of  $\beta$ - and  $\gamma$ -radiation flux densities measured in the ground atmosphere and was investigated in the work.

The idea to choose this tracer was appeared when we analyzed the results of many-years investigations of ionizing radiation fields and natural radioactivity, meteorological and atmospheric - electrical parameters (at experimental sites) in Siberia and Kamchatka were.

Numerical experiments confirmed that the ratio of the yields of beta and gamma - radiation ( $\beta/\gamma$  - index) at a certain height from the ground is constant in terms of "good weather".

The paper presents the results of studies of the reaction  $\beta/\gamma$  - index for various climatic phenomena (Fig. 1). Reported emission in the time dependence of the ratio of count rates  $\beta$ - and  $\gamma$ - pulses in the case of a cyclone passing through the territory of Japan (Fukushima) to Kamchatka (Fig. 2, 3).

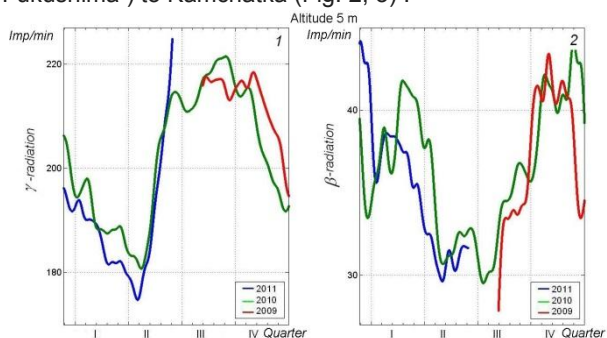


Figure 1. Dynamics of beta and gamma-radiation in the annual cycle (Paratunka, Kamchatka).

Oscillations of  $\gamma$ -radiation in Paratunka are constant until the transition of surface temperature to negative values (Fig. 1). Then  $\gamma$ -background radiation is linearly decreased until transition of surface temperature to positive values. Such change of  $\gamma$ -background is related to soil freezing and radon emission blocking. Variations of  $\beta$ -background have the global summer minimum.

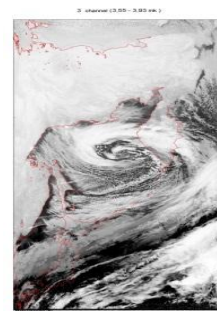


Fig. 2. Typhoon over the territory of Kamchatka (Paratunka) and Japan (Fukushima)

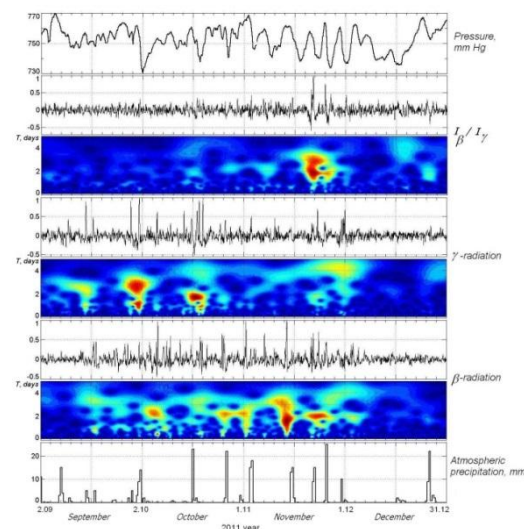


Fig. 3. Wavelet scale perturbations (days) in an electric field,  $\beta$ - and  $\gamma$ - radiation at an altitude 5 m, and normalized ratio of pulse count rate  $\beta$ - and  $\gamma$ - probes during the passage of the typhoon, downwards, respectively. Baseline data were passed through bandpass filters. Electric field: band periods is 1-30 days.  $\beta$ -,  $\gamma$ - radioactivity, and their relationship: band periods is 0,25-10 days.

Possible cause of the release have a seizure typhoon radioactive aerosols of anthropogenic origin in Japan (Fukushima).

## Acknowledgments

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# An Estimate of Canadian Population Exposure to Natural Background Terrestrial Radiation

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**Keywords:** *terrestrial radiation, exposure assessment.*

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The Geological Survey of Canada has been involved in the measurement of natural terrestrial radiation since 1934. Most publicly available airborne gamma ray spectrometric (AGRS) survey data were collected between 1970 and 2007 to support geological mapping and mineral exploration in areas of high mineral potential. Therefore, prior to the National Radon Program (NRP) there was very little coverage in populated areas. The existing data covered about 2,530,000 km<sup>2</sup> or 28% of Canada's landmass. Most coverage was flown with reconnaissance 5 km line spacing. As an important component of the NRP, AGRS surveying started in 2008 and continued to 2010. The AGRS surveys added over 200,000 line kilometres of new data covering an additional 725,000 km<sup>2</sup> in more densely populated regions. However, subject to the Canadian Aviation Regulations, AGRS surveys excluded major cities. This data gap was partially filled by truck-borne radiological surveys in a total of 21 major cities or towns near nuclear facilities across Canada from 2010 to 2014 as part of background radiation survey to support nuclear emergency responses.

In terrestrial radiation surveys, the calibration technique adopted by the Geological Survey of Canada has been to remove the effect of three sources of background radioactivity simultaneously; they are radioactivity of the aircraft and its equipment, cosmic radiation and radioactivity in the air arising from radon progeny. Environmental radiation measurements are presented as absorbed dose rates in air (nGy/h). All measurement results were determined at the nominal survey altitude (1 m above ground).

Since the aim of this study is to assess Canadian population exposure to terrestrial gamma radiation, dose calculations are performed for population centers. According to Statistics Canada, about 80% of Canadians live in urban areas where a center location can be easily be geocoded. The urban population refers to persons living in centers with a population of more than 1000 and

the area has more than 400 persons per km<sup>2</sup>. In order to cover more rural populations, communities with a population of 700 and above were considered for most provinces. In the three northern territories, communities with a population of more than 100 were included. Within the coverage of airborne and truck-borne gamma surveys, a total of 1047 communities were considered in this study which cover more than 80% of the Canadian population.

Preliminary analysis of AGRS data in 251 communities in the province of Ontario together with truck-borne measurement results in the city of Toronto showed that absorbed dose rate in air varied from 17 to 74 nGy/h with a population weighted average of 32 nGy/h. This value for terrestrial gamma radiation in Ontario is about 45% lower than the worldwide average of 59 nGy/h reported by the UNSCEAR (UNSCEAR 2000). Using 0.7 Sv/Gy as the conversion coefficient from absorbed dose rate in air to the effective dose received by adults, 0.8 as the shielding factor for indoor exposure to outdoor radiation sources, and 0.8 for the indoor occupancy fraction, the average annual effective dose in Ontario due to external exposure to natural terrestrial sources of radiation is 0.16 mSv, with 0.12 mSv related to indoor occupancy and 0.04 mSv to outdoor occupancy. The average levels for various communities in Ontario are mostly in the range 0.08 – 0.4 mSv. Similar analysis will apply to survey data available in other provinces and territories of Canada. Canadian population exposure to natural background terrestrial radiation will be assessed. Final results will be reported here.

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# Creation and Application of Voxelized Dosimetric Models: An Evaluation of Elemental Sensitivity in Dose Conversion Factors in *Apis Mellifera*

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Keywords: Voxel phantom, Organic Elemental Analysis, 3D modeling, Reference Animals and Plants  
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## Introduction

Over the past decade the International Commission on Radiological Protection has developed a comprehensive approach to environmental protection that includes the use of Reference Animals and Plants (RAPs) to assess radiological impacts on the environment. For the purposes of calculating radiation dose, the RAPs are approximated as simple shapes that contain homogeneous distributions of radionuclides (ICRP, 2008). Yet, most organisms have complex morphologies, internal structures, and densities; therefore, dose rates calculated via a homogenous model could be questioned to be unnecessarily conservative and simplified.

## 3D engineered model

*Apis Mellifera* voxel phantom model crated from 201-contoured CT planes using a general-purpose medical imaging processing software 3D Doctor



## Elemental Analysis

The materials inserted are combusted into organic matter, i.e. CO<sub>2</sub>, N-oxides and H<sub>2</sub>O in the gas phase. Then the reduction oven reduces N-Oxides to N<sub>2</sub> gas, and the gas chromatographic column separates three different peaks: N<sub>2</sub>, CO<sub>2</sub> and H<sub>2</sub>O.

Table1: Elemental composition comparison

	%N	%C	%H	%O
ICRU four component soft tissue	2.6	11.1	10.2	76.2
Honeybee soft tissue	7.6	47.6	6.9	37.8

## Dose Conversion Factors

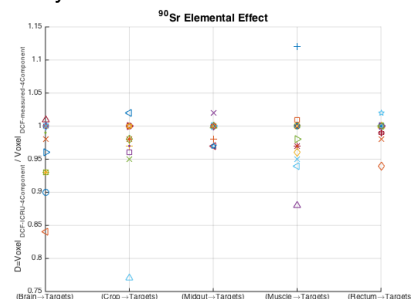
DCFs calculations to relate the concentration of a homogeneously distributed radionuclide to the dose rate in an organism as defined by the following equation:

$$DFC = \frac{\sum_i \bar{E} \times AF(\bar{E}, S \rightarrow T) \times BR \times E_c \times t_c}{C}$$

where the absorbed fractions of energy in a target tissue from a specific source region are generated by an MCNPX model.

## Results

It has been observed a significant difference in organic material composition and an elemental effect shown to be  $\pm 10\%$  on average for the selected radionuclides. In addition, the results in this study agree with other studies (Caffrey, 2012; Ruedig et al. 2015). where relatively large species are used, where increasing the level of detail does very little improvement; however, it is shown in the data of this research that at a smaller scale the importance of detail could have a slightly more significant impact in reducing the uncertainty of the calculations.



## Acknowledgments

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# Radon studies in five show karstic caves from Romania

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Keywords: radon, caves, seasonal, diurnal.

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Interest for radon in caves, originally appeared as a curiosity, and later increased due to radiological safety reasons, firstly for workers (mainly guides) who spend most of their time in this environment (Vaupotic, 2008). Moreover there are now attempts to use this gas in studies which analyse the movement of air masses inside as well as how these caves breathe (Kobal *et al*, 1988 and Vaupotic *et al*, 2001). Show caves such as Postojna (Gregoric *et al* 2001), Lascaux or Altamira (Quindos *et al*, 2014) represent natural archives of artistic interests of the Paleolithic man.

In Romania were conducted over the past three years, several measurements of radon in old salt mines (Calin and Calin, 2013) in order to characterize the environment medically as well as in terms of location of facilities for research in elementary particle physics. In the last 2 years radon measurements were initiated also in touristic karst caves (Bican-Brisan *et al*, 2011).

A comprehensive study of radon (seasonal and diurnal variations) using CR-39 detectors and devices for continuous measurements (Radim and Radon Scout) was initiated since last year. This study addresses five show caves in the West, South and Central Romania (Meziad, Bears, Muierii, Polovragi and Cetatii) and one non-touristic cave (Closani), for comparison. Integrated detectors were deployed along the touristic paths, minimum 10 points of measurement for each cave.

Table 1. Winter radon activity concentration (Bqm<sup>-3</sup>).

Cave	Mean	Minim	Maxim
Cetatii	52±38	24	157
Polovragi	879±540	41	1734
Muierii	1278±681	376	3070
Bear	393±175	81	551
Meziad	112±91	21	233
Closani	447±262	164	794

Table 1 present's data recorded during the winter season showing the average value (from 8-10 measurement points) for each cave also the maximum and minimum value. For the summer season is expected higher radon concentrations. Figure 1 display a month radon continuous measurement carried out in Closani

cave from which can be observed how the outdoor temperature and rainfall influence the concentrations of radon inside the cave.

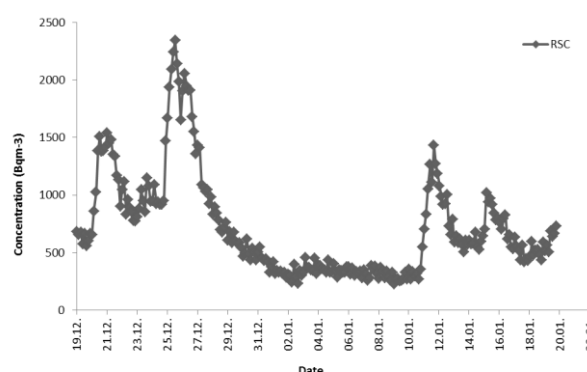


Figure 1. Continuous radon measurement in Closani cave.

## Acknowledgments

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# Analytical investigation of pottery from different Neolithic period in south-east of Albania.

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*Keywords: Energy dispersive X-ray fluorescence analysis, Archaeological pottery, Early Neolithic*

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Pottery analysis reveals information regarding the daily life and cultural aspects of the society of the ancient period. The technical skills of the ancient potters have been the subject of active research for gaining a deep insight of forgone culture.

The archaeological materials from these sites, mostly ceramics, constitute a unique case that could give information about the technological development of those societies from Early Neolithic to Eneolith.

The application of sophisticated statistical and/or multivariate methods of analysis on large experimental datasets obtained by modern analytical techniques has proven to be a useful tool for supporting archaeologists'

hypotheses regarding the age and provenance of artefacts as well as technologies and trade routes used in their manufacture and distribution.

In this paper we will consider the results of the investigation obtained by analysis using different methods like EDXRF, XRD and Optical Microscopy (OM) of territories located in the south-east of Albania from Early Neolithic to Eneolith.

The excavations have shown that in some sites the humans have been living without interruption throughout the Neolithic period

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# Distribution and seasonal variation of radioactivity, minor and tracer elements in lakes in the lignite mining area of North-Western Greece

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*Keywords: natural radioactivity; lignite region; INAA; alpha and gamma-spectrometry;*

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## Abstract

The distribution and seasonal variation of the activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in sediments and water samples in four lakes in a lignite mining area of North-Western Greece was investigated. The measurements were determined by high-resolution gamma-ray spectrometry using a HP-germanium detector in a low background configuration. In addition, Instrumental Neutron Activation Analysis (INAA) and alpha-ray spectrometry with appropriate radiochemical separations of the samples were performed, respectively. Areas with accumulation and leaching processes were estimated from the data of sediments and water samples according to the disequilibrium isotopic method for uranium (using the isotopic ratio U-234/U-238). The results are discussed taking into account several parameters such as weather conditions and hydrogeology and showed that the radiation hazard in this region is not significant.

# Application of the dilution coefficient of $^{222}\text{Rn}$ for air quality studies of $\text{PM}_{10}$ in a north-western Spanish city

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**Keywords:** radon, ceilometer, pm10, dilution

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## Objective

In this study the temporal variation of the dilution factor ( $D$ ) of atmospheric radon ( $^{222}\text{Rn}$ ) in the city of Barcelona (Spain)  $D$  has been retrieved combining atmospheric  $^{222}\text{Rn}$  concentrations data with nocturnal radiosounding and day time ceilometers data.

## Material: Atmospheric and Remote Sensing Data

In Figure 1 the hourly atmospheric  $^{222}\text{Rn}$  concentrations, obtained by continuous  $^{222}\text{Rn}$  progeny measurements carried on at the Campus Nord of the Universitat Politècnica de Barcelona (UPC) by the Institut de Tecniques Energeticas (INTE), and the hourly atmospheric concentrations of  $\text{PM}_{10}$ , available for September-October 2010 from the Marie Curie European Union framework of SAPUSS (dall'Osto et al., 2012), are presented. Remote sensing measurements on the structure of the Planetary Boundary Layer (PBL) were also measured during SAPUSS hourly, by a lidar ceilometer, and twice per day, by vertical profiles of temperature, pressure and relative humidity.

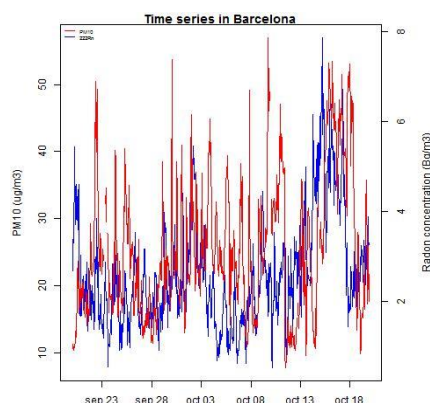


Figure 1 Atmospheric  $^{222}\text{Rn}$  (blue line) and  $\text{PM}_{10}$  (red line) measured in Barcelona in Autumn 2010.

## Method of the Dilution Coefficient (DCM)

The DCM is based on the main assumption of describing the lower atmospheric boundary layer as a box model with a finite difference approximation, implemented by Griffith et al., 2013:

$$\frac{C(t_i) - C(t_{i-1})}{dt} = F(t_i) \cdot \frac{1}{h(t_i)} - \lambda C(t_{i-1}) - D(t_i)$$
 , where  $C$  atmospheric  $^{222}\text{Rn}$  concentrations ( $\text{Bq m}^{-3}$ );  $F$  radon flux ( $\text{Bq m}^{-2}\text{h}^{-1}$ );  $h$  height of the boundary layer (m);  $\lambda$  radon decay constant ( $0.0076 \text{ h}^{-1}$ ) and  $D$  radon dilution factor ( $\text{Bq m}^{-3} \text{ h}^{-1}$ ). The relation between  $D$  and measured  $\text{PM}_{10}$  concentrations is used in order to estimate the local vs remote source of this latter pollutant.

## Preliminary results

In Figure 2 the early results are shown of distribution of dilution factor of radon during the day and the night (upper panel) and the  $\text{PM}_{10}$  concentration in relation with  $D$  (lower panel). Measured  $\text{PM}_{10}$  during the day is generally of around  $15 \mu\text{g m}^{-3}$  when air masses from poor radon areas, such as the sea, are arriving at the station and increases until  $45 \mu\text{g m}^{-3}$  when  $^{222}\text{Rn}$  increases due to air masses coming from rich radon area located in the northwest of the station corresponding to the Collserola mountain area.

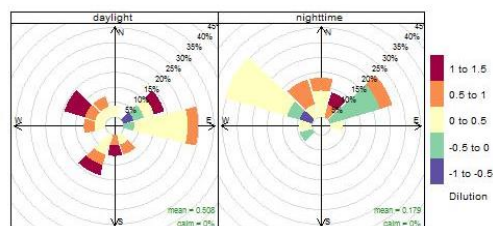


Figure 2 Polar distribution of  $^{222}\text{Rn}$  dilution factor under light time and nighttime wind conditions.

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# Comparison of several methods for thorium-isotopes determination in Environmental and Industrial Samples

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## Introduction

Thorium is a very sensitive element to matrix effects that appear at different stages of its preparation to be measured by alpha-particle spectrometry. These effects are particularly important for inorganic solid samples from environmental and industrial origin. To deal with this problem and to define the most suitable method, depending on sample type, not only from a technical point of view but also considering cost, time and chemical reagents used, a research project was started. In this work, the preliminary results of this project are shown.

## Methodology

Four experiments have been developed:

**Exp. 1:** Four different radiochemical methods, TBP+II-anionic, II-anionic, and resins type UTEVA and TEVA, have been applied to the following sample types: phosphate rock, ilmenite, sediments, sludge, ground water, blank water and reference samples of <sup>232</sup>Th and <sup>230</sup>Th. Solid samples were previously dissolved and all the methods were applied to aliquots of these dissolved samples. For ground water samples the double tracer method, with <sup>230</sup>Th and <sup>229</sup>Th, was used to analyse radiochemical and electrodeposition recoveries. So, methods were compared and one of them selected for the rest of the work.

**Exp. 2:** Four different pre-treatment methods: acid attack in a closed container, acid attack in an open container, microwave oven and alkaline fusion, have been applied to the following sample types: uranium ore, ilmenite, tioxide, soil, sludge and ashes from vegetal. In all these samples the method chosen in Step 1 was applied with the aim of assessing the best pre-treatment method, depending on the sample type.

**Exp. 3:** Chemicals that affect the electrodeposition process have been studied by chemical analysis of samples just before electrodeposition and after the radiochemical purification. The six types of samples previously described have been used and <sup>229</sup>Th tracer added after radiochemical treatment.

**Exp. 4:** Differences among the most simple lixiviation methods (EPA3050B by using both HNO<sub>3</sub> and aqua regia), and total dissolution were studied as a function of the sample type. Measurements were performed using alpha spectrometers with PIPs detectors. Uncertainties and detection limits were obtained using GUM method and ISO 11929 standard.

## Results

**Exp. 1:** All methods were comparable among them, being the II-anionic methods that supplied the highest and most reproducible results for recovery. In this method the radiochemical yield was always higher than 90%, for all type of samples, however the electrodeposition yield varied between 24 to 92%.

**Exp. 2:** Among the conclusions obtained in this Step 2, it can show that all the pre-treatment methods work properly for the samples analysed. However time, cost and chemicals involved are quite different. In all the different situations total yield was higher than 65%.

**Exp. 3:** As the application of the different radiochemical methods provided samples almost clean of chemical interferences and a multi-variant analysis shows that there is no relationship among these chemicals and the electrodeposition yield, it can be supposed that the variability of yield is more related to physical than to chemical reasons.

**Exp. 4:** For sludge, uranium ores and ashes samples, lixiviation provided results comparable to total dissolution. However, and as expected, samples like soil, ilmenite and tioxide needs total dissolution methods. With lixiviation methods, only 20% of Th is extracted from tioxide samples, 30% from ilmenite samples and around 50% from soil samples.

## Conclusions

Preliminary results show that the weakest points of Th determination methods by alpha spectrometry are the sample dissolution and the electrodeposition processes. However, analysed radiochemical methods seem to be powerful enough to deal with the samples analysed. An analysis of time involved, cost and environmental impact is being carried out. Results will be shown in the final version of this work.

## Acknowledgments

This work was supported by the Spanish Nuclear Safety Council (CSN). Special thanks to research teams from the different Universities involved in this project.

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## Public exposure to environmental radioactivity in Bilbao (Spain)

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The knowledge of radionuclides activity in air, drinking water, food and soil is necessary to assess public exposure. Hence, each Member State of the EU shall set up facilities for the permanent control of the level of radioactivity in the environment, under Article 35 of the Euratom Treaty (EU, 2010). In Spain, the entity that controls and monitors the radioactivity in the environment is the Nuclear Safety Council, which set up the National Surveillance Network.

In this context, the purpose of this work is to analyse activity concentrations in constituents of the environment that contribute to the exposure of population: air, aerosols, drinking water, diet and soil samples; in order to assess public exposure in Bilbao (located in the North of Spain) in terms of annual effective dose.

Aerosol sampling is carried out in continual by an aerosol sampling station and its filter is removed once a week. On the other hand, drinking water, diet and soil samples are collected once a month, a trimester and a year, respectively. All these samples are analysed at the laboratory by different measurement techniques (gamma and alpha spectrometry, alpha/beta counting and liquid scintillation counting), which enable us to quantify activity concentration of natural and anthropogenic radionuclides.

This way, it has been found that, for example, activity concentration of  $^{90}\text{Sr}$  in water is around  $4.6 \text{ Bq m}^{-3}$  and that of  $^{40}\text{K}$  is around  $178 \text{ Bq m}^{-3}$  while daily activity intake, due to diet, of  $^{90}\text{Sr}$  is around  $0.04 \text{ Bq d}^{-1}$  and that of  $^{40}\text{K}$  is around  $79 \text{ Bq d}^{-1}$ .

The evolution of activity concentration has also been analysed from 2000 to 2014.

Radon activity concentration is determined *in situ* by an Automatic Station. However, to determine activity concentrations of short-lived decay products of radon in air at the moment they are inhaled an analytical method had to be developed (Herranz *et al*, 2014). This method is based on a set of early gamma-ray measurements, Bateman equations and some approaches.

Additionally, in the aerosol sample with the highest activity, radiochemical isolations of U, Th, Ra, Pb and Po were carried out and their radionuclides, alpha and beta emitters, were measured by the corresponding techniques.

Then, the effective dose to the public in Bilbao is estimated taking into account internal via of exposure (aerosols, radon and thoron inhalation and drinking water and diet ingestion) and the external via (exposure from the soil) assuming a worst possible scenario. In the aforesaid scenario, outdoor occupancy factor is 1 and committed effective dose coefficients for inhaled

radionuclides are the highest among those published for slow, moderate and fast lung absorption (ICRP, 1995).

The effective dose due to inhaled aerosols and ingestion is estimated for each age group established in ICRP Publication 72 (ICRP, 1995) and *per capita*. The effective dose due to inhaled radon and thoron is estimated as UNSCEAR establishes (UNSCEAR, 2000). Finally, the external effective dose is estimated using effective dose coefficients from the FGR 12 (US EPA, 1994) for exposure from soil contaminated to a depth of 5 cm.

Thus, obtained overall average value for the effective dose *per capita* is around  $1 \text{ mSv y}^{-1}$ , 74% of which is due to the internal radiation exposure, mainly radon inhalation and diet ingestion.

In both cases, ingestion and external exposures, the major contributor to the dose is  $^{40}\text{K}$ . However, in the inhalation pathway the major contributor to the dose is  $^{214}\text{Pb}$ , which highlights the importance of determining activity concentrations of short-lived decay products in air at the moment they are inhaled and justify the development of the analytical method.

On the other hand, the contribution to the dose from anthropogenic radionuclides is around 0.08%, much lower than the contribution of natural radionuclides, and mainly due to ingestion.

### Acknowledgments

Authors acknowledge the Spanish Nuclear Safety Council as the measurements done were carried out in the framework of the European Sparse Network for environmental radioactivity monitoring.

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## Seasonal variations of radiation tracers in the ground atmosphere

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The aim of this work was search of new more informative radiation tracers of dangerous natural and man-made phenomena. Following tracers were addressed - ionizing radiation flux ratio  $\alpha/\gamma$ ,  $\beta/\gamma$ ,  $\beta/\alpha$  measured in the surface atmosphere. The objective of the work was also reasearch of seasonal variations of these tracers. Monitoring of ionizing radiation flux was produced by scintillation detectors (ATOMTEX, Belarus) in the Tomsk Observatory radioactivity and ionizing radiation (TORIR), Tomsk, Russia in the period 2009-2014.

Soil air exchange with the surface atmosphere leads to removal of soil and radioactive gases radon and thoron in atmosphere surface layer which form daughter products of decay - radioactive aerosols, during radioactive decay. Factors that manage to dynamics of gases and aerosols in the atmosphere surface layer are seasonal variations of meteorological variables - the presence of snow cover, state and change of the geological environment.

In seismically dangerous areas "background" variations due to weather and

other reasons that are not associated with the processes of earthquake preparation are superimposed on the spatio-temporal variations of the offered  $\alpha/\gamma$ ,  $\beta/\gamma$ ,  $\beta/\alpha$  tracers that are associated with changes in lithosphere stresses. These "background" variations are disturbances in terms of the search of earthquake precursors. The spectrum of these disturbances and the factors that control them are poorly researched.

Investigation of these tracers showed that the dominant period in the annual cycle is a half-year period. A typical example illustrating half-year variations of  $\alpha/\gamma$ ,  $\beta/\gamma$ ,  $\beta/\alpha$  tracers is shown in Figure 1.

There is discussion of mutual relations between the variations of  $\alpha/\gamma$ ,  $\beta/\gamma$ ,  $\beta/\alpha$  tracers, meteorological variables and presence or absence of snow cover.

### Acknowledgments

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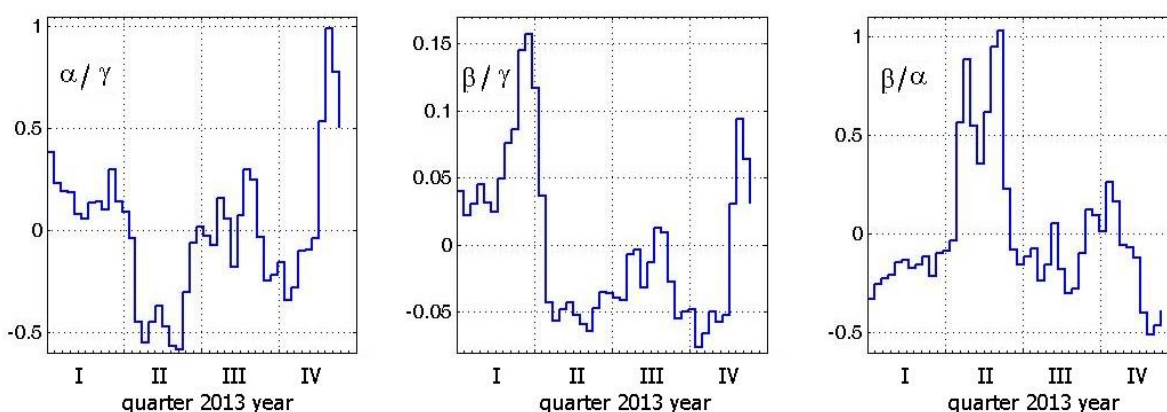


Figure 1. Half-year variations of  $\alpha/\gamma$ ,  $\beta/\gamma$ ,  $\beta/\alpha$  tracers.

# Use of reference materials RGU1 and RGTh1 in environmental gamma spectrometry measurements

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**Keywords:** Reference material, RGU1, RGTh1, self-absorption, gamma spectrometry

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High resolution gamma spectrometry is a versatile non-destructive radiometric technique that makes simultaneous determination of several radionuclides possible with little sample preparation. One key process when using this technique is the photopeak efficiency calibration and for that aims, specially when working with <sup>238</sup>U and <sup>232</sup>Th series radionuclides, the reference material RGU-1 and RGTh-1 from IAEA becomes very useful matrices. However, some methodological cautions should be taken into account with them, mainly associated to self-absorption effects.

In this work, elemental composition of both reference materials has been measured via X-Ray Fluorescence (XRF) and compared with previous data. Later, the total attenuation coefficients were assessed and some relevant conclusions are pointed out regarding the use of these reference materials. Gamma measurements were performed using an extended Range Ge detector (XtRa) in the energy range 46.5-2614.5 keV covering radionuclides from both <sup>238</sup>U and <sup>232</sup>Th natural series.

RGU-1 is compound of 0.568% in weight by an U-ore (BL-5, containing 7.09% of U and very small amount of Th) and 99.432% of silica sand. Our XRF analysis reported 50.29% Si, 47.96% O, 0.14% Al, 0.08% Fe, 0.05% Ca, 0.05% U, and others trace elements as K, Na, Ti, S.

RGTh-1, is compound of 2.843% in weight by Th-ore (OKA-2) and 97.107% of silica sand. The Th-ore contains 2.893% of Th and some amount of U (133 times lower concentration than Th). Our XRF analysis reported 50.80% Si, 46.78% O, 0.47% Ca, 0.31% Ce, 0.18% P, 0.17% Fe, 0.16% Nd, 0.13% La, 0.10% Th and another trace elements.

With this elementary composition and using the XCOM [Berger et al., 1998] database, we got the total mass attenuation coefficient [ $\text{cm}^2 \cdot \text{g}^{-1}$ ] to every material. Multiplying by their apparent density ( $1.32 \pm 0.02 \text{ g} \cdot \text{cm}^{-3}$ ) we get the Figure 1. It can be understood from his information why, having mainly the same composition (and only differing in traces), <sup>210</sup>Pb (46.5 keV) or <sup>234</sup>Th (63.3 keV) efficiencies are different between both matrices even having the same geometry and density. The self-absorption effect takes into account these small differences in composition. These differences vanish from above 200 keV.

One great advantage of using these reference materials is that no fitting of efficiency curves are

needed when working with natural radionuclides. Neither summing cascade correction to be applied, because the same radionuclides are present in the sample and in the reference material (this is not the case if the efficiency curves are constructed using artificial radionuclides).

As an example, a sediment (apparent density  $1.23 \pm 0.04 \text{ g} \cdot \text{cm}^{-3}$ ) was analysed via XRF finding in its composition mainly  $\text{SiO}_2$  but also Al, Fe, Mn, Ca... Really huge differences arise due to self-absorption effects with origin in small difference composition (Figure 1).

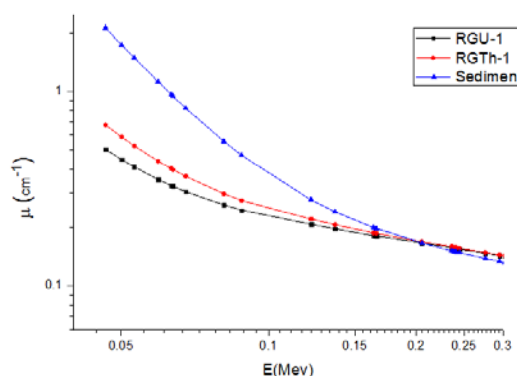


Figure 1. Total attenuation coefficient versus energy (in Log-Log scale).

The main conclusion in this work points out that below 200 keV it's completely necessary the use of self-absorption correction. Apart from that, no other correction effect is needed to be performed if we work with these reference material in soil/sediment samples with natural radionuclides via gamma spectrometry.

## Acknowledgments

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XCOM: Photon Cross Sections Database  
<http://physics.nist.gov/PhysRefData/Xcom/Text/XCOM.htm>

# Radioactive characterization of leachates and efflorescences in a phosphogypsum disposal site as a preliminary step before its restoration

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Keywords: efflorescences, phosphogypsum piles, environmental pollution, leaching waters.

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After the recent closure of the phosphoric acid plants located in the South-West of Spain (December 2010), it has been decided to restore a big extension (more than six hundred hectares) of stacks surface, where about 50 million tons of phosphogypsum (PG), the main by-product generated by these plants, had been disposed of. This PG is characterized by its high activity concentrations of several radionuclides from the uranium series, mainly <sup>226</sup>Ra, <sup>230</sup>Th, <sup>210</sup>Pb, and <sup>210</sup>Po and, to a lesser extent, U-isotopes.

The PG disposal area can be considered as a potential source of radionuclides into their nearby environment, through the waters which percolate from them and through the efflorescences formed in their surroundings and onto the surface of the piles. For this reason, a detailed radioactive characterization of the mentioned waters and efflorescences has been considered essential for a proper planning of the restoration tasks to be applied in the near future in the zone. To this end, U-isotopes, <sup>234</sup>Th, <sup>230</sup>Th, <sup>226</sup>Ra, <sup>210</sup>Pb and <sup>210</sup>Po activity concentrations have been determined by applying both alpha-particle and gamma-ray spectrometric techniques to selected water and efflorescence aliquots collected in the area.

Table 1. Activity concentrations (Bq L<sup>-1</sup>) in water samples.

	Mean	Interval	Tinto river
<sup>238</sup> U	118 ± 29	329 – 0.27	22·10 <sup>-3</sup>
<sup>210</sup> Po	17 ± 3	40 – 1.7	1·10 <sup>-3</sup>
<sup>230</sup> Th	3.6 ± 2.0	23 – 0.02	< 0.1·10 <sup>-3</sup>
<sup>232</sup> Th	0.08 ± 0.02	0.3 – 0.002	< 0.01·10 <sup>-3</sup>

Table 2. Activity concentrations (Bq kg<sup>-1</sup>) in 28 efflorescence samples.

	Mean	Interval	PG
<sup>238</sup> U	1657 ± 444	8784 – 13	220 ± 170
<sup>226</sup> Ra	402 ± 96	2186 – 25	670 ± 50
<sup>210</sup> Pb	5886 ± 2532	71133 – 105	520 ± 60
<sup>210</sup> Po	3106 ± 1065	28986 – 96	532 ± 94

The acidic waters discharging from the phosphogypsum piles into the surrounding environment

contain very high concentrations of radionuclides from the uranium series, (3-4 orders of magnitude higher than sea water or Tinto river), particularly of the U-isotopes (Table 1). On the contrary, the activity concentrations in the efflorescences cover an ample range, with generally very high activity concentrations of <sup>210</sup>Pb and <sup>210</sup>Po (Table 2), which are, in turn, higher than other typical radionuclides of phosphogypsum, such as <sup>226</sup>Ra and <sup>238</sup>U. These facts clearly highlight the different trend of the radionuclides of the uranium series in the different processes occurring in the area, that affect their behaviour in terms of dissolution in the acidic waters, incorporation to the efflorescences during their formation, etc.

However, in spite of the general high activity concentrations determined both in draining waters and in efflorescences, the relatively low weight of the outflows from the piles in comparison with the water flow in the estuary, and the low density of existing efflorescences imply a moderate radioactive environmental impact of the disposal area on the estuary as a whole, although this impact is more significant in the surrounding salt-marshes. As a consequence, in the planned restoration, and in an effort, from the radioactive point of view, to return as much as possible to the situation existing before the disposal of PG in the estuary was started, two main actions should be taken: the outputs of acidic waters from the disposal area should be banned, and the existing efflorescences should be removed and properly managed.

## Acknowledgments

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# Ionization chamber of new conception for environmental gas radon measurements

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**Keywords:** radon, ionization chamber, nuclear detector, radioactivity, health safety.  
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Long exposure to high Radon levels leads to an increase of developing lung cancer risk, due to irradiation of lung tissue by the  $\alpha$  particles emitted by Radon and its decay products.

Another very interesting aspect related to the Radon concentration is its potential use as a seismic events precursor, according to some geophysical models that suggest the release of Radon from the underground as a result of tectonic deformation responsible for the earthquake.

This paper shows the study of a gas detector running in open air, characterized by a metal cylindrical cathode and a filiform anode placed along the axis of the cylinder.

First the gas chamber behavior was studied in presence of a solid  $^{241}\text{Am}$  source in order to provide a efficiency evaluation at energy range of interest and to identify the tension values of the ionization chamber operating mode.

Next the detector characterization was performed directly with Radon gas source by using a pumping system in order to allow the gas to be distributed along the entire active volume.

The resulting tests will be presented by focusing on the absolute efficiency evaluation, Minimum Detectable Activity (MDA) and time response to radioactivity.

The obtained results made it possible to quantify system performances useful for radon measurements employment.

Moreover, on the basis of these results, an updated version of the detector was built and characterized: the cathode was replaced by a cylindrical steel plate, with a perforated mesh structure. In this way the detecting performances were almost the same (the electric field seen by the alpha particle is not significantly influenced) and a direct

operation into the atmosphere, without using of any pump system was allowed, thus reducing weight, size and energy consumption.

The main advantages of using this detector are: quick response, strength, compactness, cheapness, long enough autonomy of operation (a few months), easy to install and use.

Hence this detector could be potentially used in a widespread network on the territory for the Radon gas concentration measurement in living environments and/or aimed at testing of predicting earthquakes theoretical models.

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# Vertical Profile of $^{137}\text{Cs}$ , $^{210}\text{Pb}$ and $^{40}\text{K}$ in Ghardia Region, Algeria

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**Keywords:** Sahara desert, nuclear test, nuclear accident, soil profile.

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The depth profiles of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  concentrations in two types of soil from different areas of Algeria were examined.

The soil collection areas are near the location where the French nuclear tests took place at 1960 – 1961. The collection area of sample 1 is rocky with sand while the collection area of sample 2 is porous with stones and sand.

Samples collected using geological carrot descending 2, 5 and 10 cm each time. Sampling depth: 0 – 70 cm. The Sand grain size was  $\sim 0.4$  mm.

Soil Sample 1 collected 20 km north from the Ghardia region and characterized as rocky with sand.

Soils Sample 2 collected about 70 km northern from the collection place of soil sample 1 and characterized as porous with rocks and sand.

The measurements of the soil samples were carried out by gamma spectroscopy using high resolution detectors of low background. For  $^{210}\text{Pb}$  (46.5 keV) a Ge planar detector have been used, with active area 2000 mm<sup>2</sup>, thickness 20 mm, energy resolution (FWHM) 400 eV at 5.9 keV or 700 eV at 122 keV. While for  $^{137}\text{Cs}$  (661.65 keV) and  $^{40}\text{K}$  (1460.74 keV) a 20% Efficiency HPGe Low background, high resolution 1.86 keV at 1.33 MeV. Results are given in Table 1.

Table 1. Maximum and minimum values of the activity of the two samples

	Sample 1	Sample 2
$^{210}\text{Pb}$	max: $\sim 50$ Bq/kg min: $\sim 27$ Bq/kg	max: $\sim 75$ Bq/kg min: $\sim 37$ Bq/kg
$^{137}\text{Cs}$	max: $\sim 3$ Bq/kg min: $\sim 0.2$ Bq/kg	max: $\sim 6$ Bq/kg min: $\sim 4$ Bq/kg
$^{40}\text{K}$	max: $\sim 90$ Bq/kg min: $\sim 75$ Bq/kg	max: $\sim 180$ Bq/kg min: $\sim 140$ Bq/kg

The three radionuclides have different rhythm of diffusion and different transfer rate at the two different soils. The diffusion and transport of radionuclides in the two soils are different and depend on the natural soil composition. Sample 1 shows uniformity in the distribution of radionuclides, without a clear maximum (peak), while sample 2 shows a clear peak at 20 – 50 cm depth which is probably due to the different nature of the soil samples. The  $^{210}\text{Pb}$  activity values ranged between 27 Bq kg<sup>-1</sup> and 50 Bq kg<sup>-1</sup> in sample 1 and between 37 Bq kg<sup>-1</sup> and 75 Bq kg<sup>-1</sup> in sample 2, respectively. The  $^{137}\text{Cs}$  activity values ranged between 0.2 Bq kg<sup>-1</sup> and 3 Bq kg<sup>-1</sup> in sample 1 and between 4 Bq kg<sup>-1</sup> and 6 Bq kg<sup>-1</sup> in sample 2. The  $^{40}\text{K}$  activity values ranged between 75

Bq kg<sup>-1</sup> and 90 Bq kg<sup>-1</sup> in sample 1 and between 140 Bq kg<sup>-1</sup> and 180 Bq kg<sup>-1</sup> in sample 2. The similar behavior of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  at both soil samples is probably due to the similar chemical behavior.

The analysis of samples showed that:

$^{137}\text{Cs}$  concentration values in all samples were lower than most  $^{137}\text{Cs}$  values observed in Europe.

Similar vertical distribution profiles of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  have been observed at both two different samples

Concentration values of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  at sample 1 are lower than their corresponding values in sample 2

In sample 1 was observed uniform distribution of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  at all layers.

In sample 2, a clear maximum of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  concentration value was observed between the depth of 20 – 50 cm

The most probable explanation is that there is an easier movement of radionuclides in the soil – porous with stones and sand.

These observations can be used in existing national or international data bases for providing significant information for site characterization of potential contaminated areas.

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# An arrangement for the study of distribution coefficients in soils naturally enriched in uranium isotopes and $^{226}\text{Ra}$

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**Keywords:** distribution coefficient, transfer, soil, plant, uranium, radium.

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Although the transfer of radionuclides from the soil to plants is a complex process, most dosimetric models involving the two compartments make very simple approximations. Equilibrium models are based on instantaneous linear relationships between the concentrations in the plant and in the soil, and dynamic models assume constant uptake rates. Despite this simplicity, the uncertainties associated with the results of these models are often large (IAEA, 2010).

The general transfer process, beginning with the soil as a repository of radionuclides or heavy metals, can be divided into a few steps, the first of which will involve just the soil itself. In this sense, before introducing the plant and its biochemical activity to determine the Transfer Factor ( $TF$ ), one needs to understand the processes that establish the availability of those elements (Higley and Bytwerk, 2007). This approach will reduce the large uncertainties commonly associated with the  $TF$  derived from an evaluation of the distribution coefficient,  $K_d$ .

The availability of a radionuclide for its assimilation by the plant depends on its desorption from the solid phase in contact with the soil solution, and is influenced by numerous physical, chemical, biological, and environmental factors (Tiensing *et al.*, 2001). Only a part of the available fraction will be finally incorporated into the plant (Tyler, 2000).

In a natural scenario, the soil acts as a buffer attenuating the effects of the external agents that disrupt the trend towards a state of equilibrium. Aside from the influence of the biota, which is of great importance but also very complex, the soil's characteristics help one to understand its role in the trend towards equilibrium. All of its physical characteristics (structure and particle size, hydrodynamic behaviour, etc.), its chemical and mineralogical composition, and even environmental parameters, as a block will determine the radionuclides' distribution coefficients ( $K_d$ ) between the solid matrix and the soil solution (Raguz *et al.*, 2013).

In the present study, different experiments were designed to determine the available fraction of the natural uranium isotopes and  $^{226}\text{Ra}$ , and the role the soil's physical and chemical characteristics play in that availability.

Extracts of the soil solution were obtained by centrifugation, using a high-performance centrifuge and custom-designed devices to adapt the fixed-angle rotors to the requirements of the experiments. The experiments were planned with varied initial conditions of moisture, incubation period, and effective tension applied to the

soil. Figure 1 is a schematic diagram of the factorial study.

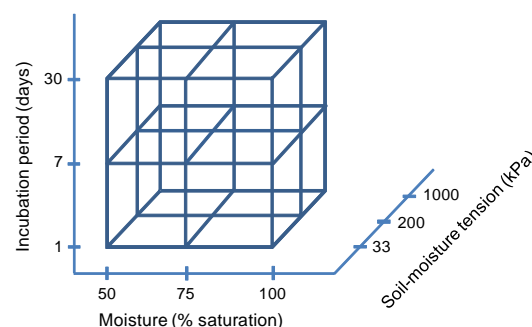


Figure 1. The  $3^3$  factorial design.

The analyses were performed on three physical fractions of the soil with different particle size distributions, a parameter that is well recognized as having a major influence on radionuclide retention and release.

The centrifuge was also used to characterize the soil hydromechanically. Van Genuchten's model was fitted to the experimental results of retained moisture versus the effective matric potential varied by means of the rotor's angular velocity. This curve allows one to establish the different origins of the soil solution in terms of the forces that link the water to the soil matrix (gravitational water and capillary water).

## Acknowledgments

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# The Uranium Isotope in the Characterization of Groundwater in Anthemountas River Basin, Northern Greece

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Keywords: groundwater,  $^{234}\text{U}/^{238}\text{U}$ , alpha-spectrometry, electro-deposition, radio-separation

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The activity concentrations of  $^{238}\text{U}$  and  $^{234}\text{U}$  have been determined in groundwater samples in Anthemountas river basin, Northern Greece. The analysis was performed by alpha spectroscopy after pre-concentration and separation of uranium by cation exchange and finally its electro-deposition on stainless steel discs. The obtained isotopic ratio  $^{234}\text{U}/^{238}\text{U}$  varies between 1.05 and 3.50 and is correlated with the different aquifer types and water flow paths in the study area, with the observed low  $^{234}\text{U}/^{238}\text{U}$  activity ratio values to indicate most probably new-type waters.

In the case of Anthemountas river basin the available data revealed a very complex system of aquifers with significant extent in both lateral and vertical sense. Based on the geological structure, three types of aquifers are occurred: porous, karstic and fissured rocks aquifer (Kazakis et al. 2013). Karstic (limestones) aquifer discharges groundwater through fault springs which is mixed with hydrothermal waters. Fissured rock aquifers are developed in Gneiss, Schists, Granodiorite, Gabbro, Dunites, Peridotites, locally covered by sediments and they are mainly elongated in fault zones. In the alluvial sediments (neogene and terrace system) the porous aquifer is developed, located in the lowlands of the basin and consists of gravels, conglomerates, sands and sand with clay. The porous aquifer is divided vertically in the upper shallow unconfined layer (mean thickness of 80 m) and the underline confined layer which thickness is up to 300 m. Hydrothermal waters are hosted in underline sandstones and limestone in depths greater than 300 m. Groundwater of the basin is the main supply source for domestic, industrial, livestock and irrigation uses and consequently its hydro chemical composition is of outmost importance.

The separation of the alpha radionuclides from the aqueous matrix was performed by cation exchange using Chelex-100 resin (Konstantinou et al. 2004, Ioannidou et al. 2011, Antoniou et al. 2008)]. The analysis was performed by alpha spectroscopy (using PIPS detectors) for 25 hours. The minimum detection activity of the method (95% confidence level) is 1.2 mBq  $^{238}\text{U}$  L<sup>-1</sup> corresponding to 97 ng  $^{238}\text{U}$  L<sup>-1</sup>.

According to the preliminary experimental results (Table 1) the uranium concentration in the region of study varies strongly between 1.7 and 14.5  $\mu\text{g}$  L<sup>-1</sup>. Uranium concentration in natural waters is affected by a number of factors, such as the lithostratigraphic formations, their mineral content, the chemical behavior of the nuclide, the origin of a groundwater etc.  $^{238}\text{U}$  and

$^{234}\text{U}$  activity concentrations ranged between 21.0-179.09 mBq L<sup>-1</sup> and 45.2-197.0 mBq L<sup>-1</sup> respectively. The obtained isotopic ratio  $^{234}\text{U}/^{238}\text{U}$  varies between 1.05 and 3.5 and in some samples is much lower than indicated in literature.

Table 1. Uranium concentrations in groundwater samples during winter 2013

a/a	pH	$\mu\text{S}/\text{cm}$	$^{238}\text{U}$ (mBq/L)	$^{234}\text{U}$ (mBq/L)	$^{234}\text{U}/^{238}\text{U}$	U (ppb)
No1	7.90	400	40.80	61.20	1.50	3.3
No2	7.60	423	61.74	117.30	1.90	5.00
No3	7.70	439	43.05	45.20	1.05	3.49
No4	7.90	517	24.63	50.50	2.05	1.99
No5	7.20	1550	21.00	73.50	3.50	1.70
No6	6.60	1277	179.09	197.0	1.10	14.50
No7	8.10	611	44.20	66.30	1.50	3.58
No8	7.90	771	61.18	104.00	1.70	4.72

The observed low  $^{234}\text{U}/^{238}\text{U}$  activity ratio values indicate most probably new-type waters with a stronger contribution of a local recharge component to the groundwater (infiltration of rainwater). Lower values were observed in the shallow porous aquifer and the fissured rock aquifers due to the high recharge rate. In contrast, the deeper porous aquifer (in spring No\_5) and the wells with mixed fresh and hydrothermal water have higher isotopic ratio ( $^{234}\text{U}/^{238}\text{U}$ ).

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# Influence of precipitation on fluxes of alpha, beta and gamma radiation in the ground atmosphere

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Keywords: atmospheric radiation, ground atmosphere, thunderstorm, precipitation.

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The aim of this work was to study consistency of changes in the intensity of  $\alpha$ -,  $\beta$ - and  $\gamma$ -radiation in the ground atmosphere during the passage of cyclones and thunderstorms in the summer and winter periods.

About 100 days with precipitation greater than 5 mm had been recorded during the summers of 2011-2013. Variations in flows of  $\alpha$ -,  $\beta$ - and  $\gamma$ -radiation were obtained during clear weather, lightning storms and precipitation.

Example of  $\alpha$ -,  $\beta$ -,  $\gamma$ - radiation flux variations and their relationship during a series of winter storms is shown in Fig. 1. In the same figure the variation of the electric field and the polar electrical conductivities are shown.

The relationship between the increases in the flux of  $\alpha$ -,  $\beta$ -,  $\gamma$ - radiation, atmospheric electricity, meteorological variables and precipitation are discussed.

## Acknowledgments

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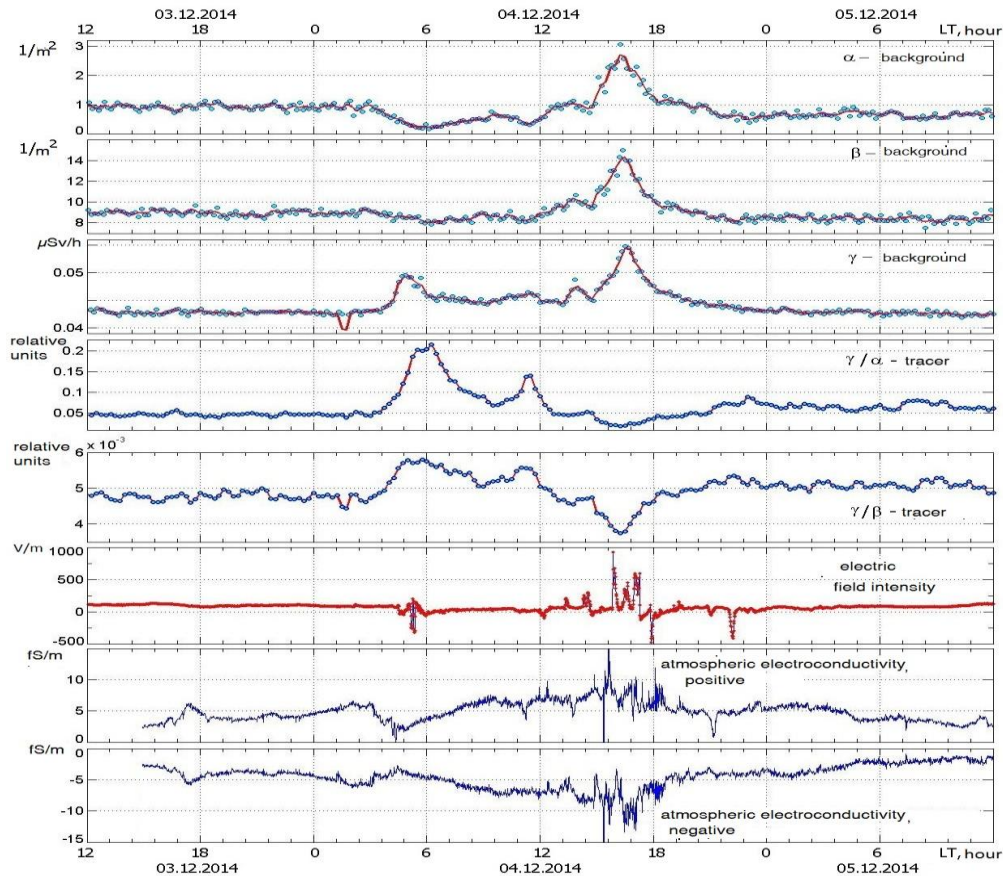


Figure 1.  $\alpha$ -,  $\beta$ - and  $\gamma$ - radiation flux variations and their relationship during a series of winter storms

# Radon indoor concentration and its seasonal variation in Athens, Greece

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Keywords: radon, CR-39 detectors, seasonal variation  
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Radon ( $^{222}\text{Rn}$ ) and its daughter products inhalation represents a major source of natural radiation exposure and according to WHO (2009) radon is the second most important cause of lung cancer after smoking.

The study of the indoor radon concentrations in Greece represents an area of great interest. Until now, there were performed two large surveys: one by the University of Athens (1995-1998) and one from 1999 to 2006 by the Nuclear technology laboratory, Aristotle University of Thessaloniki (Clouvas *et al*, 2009).

In the light of the above, we performed an indoor radon survey in 25 dwellings (apartments and houses) in Athens over three seasons from 2013 to 2014. The aim of this study is to contribute with new data on both the indoor radon concentration and on its seasonal variation.

The experimental design consisted in exposing the CR-39 solid state nuclear tracks detectors for 3 periods of three months from March 2013 to March 2014. The detectors were placed at a distance of at least 0.5 m from the wall and, in order to obtain more representative samples of air from the breathing zone, as far away from the windows and doors. The measurement protocol was performed as described by Cosma *et al* (2009). The statistical analysis was performed using GraphPad Prism (version 6.01).

In table 1 are presented the results regarding the range of radon indoor concentrations during the three seasons. The percent of the houses which showed a radon indoor concentration higher than  $100 \text{ Bq/m}^3$  was about 8% during the spring, while throughout the summer the percent increases up to 36%, and in the winter was recorded the highest increase, up to 56%, and one of the houses exhibited a concentration higher than  $200 \text{ Bq/m}^3$ .

Table 1. Radon concentration during three seasons

Season	Radon concentration ( $\text{Bq/m}^3$ )
March-June 2013	25-189
June-September 2013	42-186
January-March 2014	79-245

Moreover, there is observed a seasonal variation of radon concentration (Figure 1). The highest mean average of indoor radon concentration was recorded during the winter season ( $137.5 \text{ Bq/m}^3$ ), followed by the summer season ( $96.10 \text{ Bq/m}^3$ ), while the lowest one was noticed during the spring season ( $54 \text{ Bq/m}^3$ ).

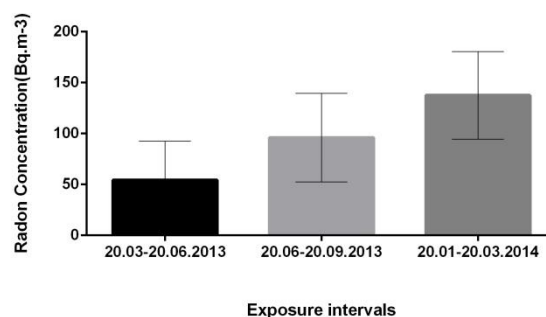


Figure 1. Radon indoor concentration mean average ((Each bar represents Mean  $\pm$  SD [Standard deviation] for n= 25 samples)

The main conclusions of our study can be resumed as follows:

- houses located on ground floor show a higher concentration of radon,
- building insulation reduces the indoor radon concentration,
- houses occasionally used by owners' exhibit a higher concentration of radon,
- the increase of radon inside the houses can be also attributed to the fact that most air conditioning devices recycle the indoor air, thus blocking the radon indoor.

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# Potassium-40 in the Coastal Area of Montenegro – dose rate assessment

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Keywords: K-40, dose rates, Coast, Montenegro.

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The Coastal region of Montenegro adjacent to the South Adriatic Sea makes ~11.5 % of the total country area, as well as ~24 % of the total population. The present study considers dose rates due to natural radionuclide <sup>40</sup>K in its different compartments – soil, sand, sediment, seagrass (*Posidonia oceanica*), seawater and fish (*Mugil cephalus*). Since terrestrial radiation mainly depends on geological aspects, an analysis of the region geology was also performed, i.e., characteristics of two geotectonic units – Budva-Cukali and Adriatic-Ionian, are also given, as well as typical soil types.

The top 5 cm uncultivated soil from 10 locations, sand from 12 renowned beaches, seawater from 8 locations (Fig. 1), sediment, 2 seagrass samples, 10 whole fish and 2 muscles, were measured using gamma spectrometry – multidetector spectrometer PRIPYAT-2M (characteristics can be seen from refs. Antovic and Svrkota, 2009; Antovic and Antovic, 2011), and the ORTEC HPGe spectrometers (GEM-40190, 30185-S).



Figure 1. A map of sampling:  $\blacklozenge$ - soil,  $\square$ - sand,  $\circ$ - seawater.

Dose rates were inferred from <sup>40</sup>K activity concentrations using the formulas from the UNSCEAR reports (UNSCEAR, 2000; UNSCEAR, 2008). In regard to soil and sand, dose rates due to <sup>40</sup>K (from 258 to 665 Bq kg<sup>-1</sup>, and from 22.6 to 298 Bq kg<sup>-1</sup>, respectively) were found to range from 10.8 to 27.7 nGy h<sup>-1</sup>, and from 0.94 to 12.4 nGy h<sup>-1</sup>, respectively. The lowest activity concentrations in seawater were found for samples taken from 6.5-7 m depth in the Boka Kotorska Bay (Sv. Stasije and Dobrota – 2.81 and 3.31 Bq L<sup>-1</sup>, respectively), while in surface seawater – from 3.14 (Budva-Jaz) to 8.84 Bq L<sup>-1</sup> (Tivat). In sediment from the

Boka Kotorska Bay (Dobrota), 123 Bq kg<sup>-1</sup> of <sup>40</sup>K was measured, resulting in an illustrative dose rate of 5.13 nGy h<sup>-1</sup>. The results of dose assessment in biotic samples (seagrass from Sv. Stasije and Dobrota, whole *M. cephalus* and its muscles) are reported in Table 1.

Table 1. Dose rates due to <sup>40</sup>K.

Sample	K-40, Bq kg <sup>-1</sup>	Dose rate, nGy h <sup>-1</sup>
Seagrass-1	389±14	170±6
Seagrass-2	491±26	210±11
Fish-1	108±7	43.4±2.8
Fish-2	96.8±7.5	39.0±3.0
Fish-3	80.9±8.1	32.8±3.2
Fish-4	127±9	50.8±3.6
Fish-5	88.9±10.5	36.0±4.2
Fish-6	84.7±10.7	34.3±4.2
Fish-7	100±13	40.3±5.1
Fish-8	121±14	48.5±5.5
Fish-9	74.3±5.1	30.3±2.1
Fish-10	86.7±5.5	35.1±2.2
Muscle-9	116±11	46.5±4.3
Muscle-10	120±12	48.1±4.7

For a comparison, the results for fish at Trombay, Mumbai, India (Singhal *et al*, 2009), for example, where in seawater <sup>40</sup>K was found to be from 4 to 7 Bq L<sup>-1</sup> (i.e., comparable with the level on the Coast of Montenegro), in bottom sediment – 80-150 Bq kg<sup>-1</sup>, in fish 150-200 Bq kg<sup>-1</sup> (i.e., higher than in *M. cephalus*), showed exposure due to <sup>40</sup>K in the range from 525 to 700  $\mu$ Gy y<sup>-1</sup>, i.e., from around 60 to around 80 nGy h<sup>-1</sup>. In the present study obtained doses to *M. cephalus* due to <sup>40</sup>K in seawater and fish themselves can be considered as lower (even the highest one is lower than minimal in fish at Trombay).

## Acknowledgments

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# A Study on the Correlation between Soil Radon Potential and Average Indoor Radon Potential in Canadian Cities

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**Keywords:** soil radon, indoor radon, radon-222.

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Radon has been identified as the second leading cause of lung cancer after tobacco smoking. Since radon in soil is believed to be the main source of radon in Canadian homes, a soil radon potential (SRP) index determined from soil gas radon concentration and soil permeability with rather simple technology can be a useful tool for prediction of the average indoor radon potential in an area and provide scientific support to future land-use planning. The SRP index is defined as,  $SRP = (C - C_0) / (-\log(P) + \log(P_0))$  where  $C$  is the radon concentration of the soil gas in  $\text{kBq/m}^3$ , and  $P$  is the soil permeability in  $\text{m}^2$ .  $C_0$  and  $P_0$  are set to  $1 \text{ kBq/m}^3$  and  $1 \cdot 10^{-10} \text{ m}^2$ , respectively.

From 2007 to 2010, SRP indexes were determined for more than 430 sites in 13 Canadian cities. Average soil gas radon concentrations were calculated at each site, generally from measurements made using 5 probes inserted to a depth of 60 to 80 cm in a 10m by 10m area. At each site, the in-situ soil gas permeability was normally measured with at least 2 probes and the average permeability determined. For each of the more than 1400 probes where in-situ soil permeability measurements were conducted, subjective descriptions of the relative difficulty of collecting soil gas with a syringe were also recorded as easy, medium or hard. This provided a semi-quantitative estimation of permeability for those sites where in-situ soil permeability measurements were not possible for logistical reasons. The SRP for a city is the average of SRPs from all sites surveyed within the city.

The indoor radon concentrations varied widely in a geographic area, and followed log-normal distribution. A log-normal distribution is a two-parameter distribution characterised with the geometric mean (GM) and geometric standard deviation (GSD), therefore, indoor radon potential of a geographic area or a city should be a quantity representing the characteristics of the two-parameter distribution of indoor radon concentrations in that area. The percentage of homes above the Canadian action level of  $200 \text{ Bq/m}^3$  was chosen as the indicator of indoor radon potential for an area. The information identifying the percentages of homes above  $200 \text{ Bq/m}^3$

in various cities was obtained from special radon surveys in cities of Ottawa (Chen et al. 2008), Winnipeg (Chen et al. 2009), Halifax and Fredericton (Chen et al. 2011), and from recently completed radon and thoron survey in 33 metropolitan areas of Canada (Chen et al. 2015).

A reasonable correlation ( $R^2=0.47$ ) between SRP and percentage of homes above  $200 \text{ Bq/m}^3$  average was found for the 13 Canadian cities studied here. For 5 cities (Ottawa, Halifax, Fredericton, Winnipeg and Regina) where detailed in-situ soil permeability was measured in more than 20 sites per city, a strong correlation ( $R^2=0.81$ ) was found. If, instead of SRP, considering the average soil radon concentration in those 5 cities, the correlation to percentage of homes above  $200 \text{ Bq/m}^3$  decreased slightly to  $R^2=0.78$ . This clearly indicates that radon in soil is the main source of radon in homes while soil permeability also plays an important role for soil radon potential and, ultimately, the indoor radon potential. The results showed that SRP can serve as a reasonable indicator for the average indoor radon potential, i.e. percentage of homes above  $200 \text{ Bq/m}^3$  in a geographic area.

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## Measurement of Natural and Artificial Radioactivity in The Lebanese Environment for Total Dose Assessment

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**Keywords:** Environmental radiation monitoring, gamma spectroscopy, alpha spectroscopy, activity concentration, annual effective dose

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The Environmental Radiation Control Department at LAEC, established a National Environmental Radiation Monitoring Program in 2009, in order to control natural and artificial radioactivity levels in various compartments of the Lebanese environment, set radiation baseline levels and detect any abnormal releases resulting from any nuclear accident. This enables to plot the trend of radioactivity over time.

Gamma emitters were analyzed periodically. uranium isotopes and  $^{210}\text{Po}$  and were analyzed in some selected samples, using alpha spectrometry after chemical separation. Two selected rivers are monitored. Three coastal cities are selected for marine sampling. Soil samples were collected from uncultivated areas. Foodstuff items representing the main diet constituents of the majority of Lebanese population (Nasreddine *et al*, 2008) are picked up from local market, such as crops, industrial processed food, meat, chicken, milk and dairy products. Air monitoring is carried out. Gamma measurements were performed in an accredited ISO 17025 laboratory using gamma spectrometers with high purity germanium detectors (El Samad *et al*, 2013). This work represents the data obtained over six years. Maximum activity concentration of  $^{137}\text{Cs}$  in soil was  $113 \pm 2.4 \text{ Bq kg}^{-1}$  with an external effective dose  $4.16 \mu\text{Sv/year}$ . While, its content in most analyzed samples was below minimum detectable activity (MDA) approximately  $0.02 \text{ Bq kg}^{-1}$ . Some species showed  $^{137}\text{Cs}$  content slightly higher than MDA, mainly one coffee brand, a milk sample, a wheat sample and a jam sample. Its concentration was  $0.50 \pm 0.07 \text{ Bq kg}^{-1}$ ,  $1.98 \pm 0.08 \text{ Bq kg}^{-1}$ ,  $1.10 \pm 0.06 \text{ Bq kg}^{-1}$  and  $2.5 \pm 0.22 \text{ Bq kg}^{-1}$  respectively. However highest value,  $5.24 \pm 0.13 \text{ Bq kg}^{-1}$ , was detected in whey powder imported from Poland.

$^{40}\text{K}$  results, the main detected natural radionuclide, varied between matrices but still comparable over years. Highest values were detected in milk powder, coffee and tea,  $646 \pm 15 \text{ Bq kg}^{-1}$ ,  $640 \pm 16 \text{ Bq kg}^{-1}$  and  $699 \pm 18 \text{ Bq kg}^{-1}$  respectively. The total annual effective dose from ingested food was based on the  $^{40}\text{K}$  intake only, as the activity concentrations of other natural radionuclides, as well as that of  $^{137}\text{Cs}$  were negligible in nearly all food samples except in fish, where  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are major contributor in internal dose. Noting that the yearly consumption rate of fish in Lebanon is the lowest one compared to other species (Nasreddine *et*

*al*, 2008). The total annual effective internal dose from 2009 till 2014 was,  $56 \mu\text{Sv y}^{-1}$ ,  $42 \mu\text{Sv y}^{-1}$ ,  $104 \mu\text{Sv y}^{-1}$ ,  $138 \mu\text{Sv y}^{-1}$ ,  $146 \mu\text{Sv y}^{-1}$ , and  $143 \mu\text{Sv y}^{-1}$ , respectively. This accretion is due to the expansion of type and number of commodities. It was found that the dietary intake of  $^{40}\text{K}$  was lower than worldwide average value  $0.3 \text{ mSv y}^{-1}$  (UNSCEAR 2000). Figure 1 represents the activity concentration of  $^{40}\text{K}$  in fruits and vegetables.

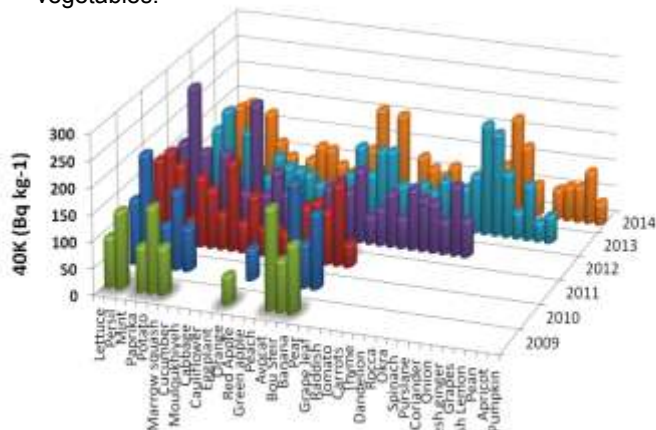


Figure 1. Activity concentration of  $^{40}\text{K}$  in fruits and vegetables

Activity concentrations of  $^{232}\text{Th}$ ,  $^{210}\text{Pb}$ , and  $^{210}\text{Po}$  in marine samples were comparable between locations. While the  $^{210}\text{Po}$  content in fish samples varied between  $43 \pm 3 \text{ Bq kg}^{-1}$  in dry mass to  $150 \pm 9 \text{ Bq kg}^{-1}$  according to species and location.  $^{234}\text{U}$  and  $^{238}\text{U}$  were analyzed in seawater, results varied from  $37 \pm 2 \text{ mBqL}^{-1}$  to  $68 \pm 5 \text{ mBqL}^{-1}$  and  $33 \pm 2 \text{ mBqL}^{-1}$  to  $57 \pm 6 \text{ mBqL}^{-1}$  respectively. After Fukushima accident, grass and rain water samples were collected. Due to wet deposition,  $^{131}\text{I}$  was detected in grass, whose activity concentration varied between  $0.4 \pm 0.03 \text{ Bq kg}^{-1}$  and  $0.9 \pm 0.1 \text{ Bq kg}^{-1}$ , as well as  $^{137}\text{Cs}$  content varied in the range of  $0.15 \pm 0.02 \text{ Bq kg}^{-1}$  to  $0.4 \pm 0.05 \text{ Bq kg}^{-1}$  in some seafood samples imported from Japan and its neighboring countries.

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# Outdoor Radon Monitoring Network to study the behaviour of $^{222}\text{Rn}$ over Spain over Spain

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**Keywords:** radon, outdoor air, natural radionuclides, Spain, atmosphere

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The noble radioactive gas  $^{222}\text{Rn}$  is widely used as tracer to improve inverse modelling techniques which use high quality and spatial density data of atmospheric concentrations of a specific gas to retrieve its fluxes over different regions. Several European monitoring network of greenhouse gases have already included the  $^{222}\text{Rn}$  measurements nevertheless there is still a lack of harmonized data over the Mediterranean area (ej. [www.ingos-infrastructure.eu](http://www.ingos-infrastructure.eu))

Different  $^{222}\text{Rn}$  ARMON (Atmospheric Radon Monitor) monitors either have been set up (Vargas et al. 2015; Grossi et al., 2014; Grossi et al. 2013), or are planning to set up in stations located in mountain as well as in coastal areas of the Spanish geography (Table 1) and with different local radon flux sources (Figure 1). The idea is to create an Outdoor Radon Monitoring Network (ORMON) starting from the collaboration between the Institut Català de Ciències del Clima (IC3), the Institut de Tècniques Energètiques (INTE) of the Universitat Politècnica de Catalunya (UPC), the Universidad de Huelva (UHU), and other organizations that like to participate. The network aims studying the atmospheric variability of radon gas in different environments and under different synoptic situations. In this work the temporal variability of outdoor  $^{222}\text{Rn}$  in Spain will be presented and analysed.

Station	Lat.	Long.	Altitude (amsl)	Type of station	Managed	Data
DEC3	40.74N	0.79E	10	NE coastal	IC3	06/2014-present
GIC3	40.22N	-5.14E	1440	Central Mountain	IC3	11/2012-present
ARE	37.10N	-6.70E	10	SW coastal	INTA-INTA	03/2012-02/2013
UHU	37.27N	-6.92E	10	SW city	UHU	07/2014-present
BCN	41.38N	2.12E	80	NE city	INTE	10/2014-present
XIC3	42.07N	-8.04E	460	NW mountain	IC3	-
SGC3	36.72N	-5.36E	980	SE mountain	IC3	-

Table 1 Characteristics of ORMON stations

## Acknowledgments

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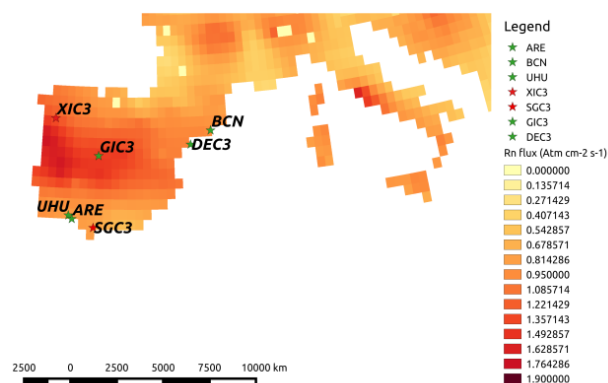


Figure 1 European radon flux map (Szegvary et al., 2009) and location of ORMON stations

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# EVALUATION OF URANIUM IN ORGANS OF RESIDENTS FROM AN URANIUM-RICH REGION USING TEETH AS BIOINDICATORS

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**Keywords:** Environmental Uranium contamination; Biokinetics model calculations; Uranium burdens in organs

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The Uranium extraction and processing plant of INB (Brazilian Nuclear Industries) is in Caetité, a city located in a region hosting the largest Uranium reserve of Brazil. The degree of Uranium contamination in the Caetité population was inferred in a previous work by using teeth as bioindicators, where a quite high uranium concentration was measured in this organ, about 160 times higher than the world-wide average. (Prado et al., 2008). However, the evaluation of radiobiological risks of populations living in contaminated areas as Caetité city, or any other contaminated locality, requires estimates of Uranium burdens in key-organs other than teeth, as the skeleton, kidneys, liver, tissues and blood. This is accomplished in this work by means of calculations with the use of the STATFLUX / ICRP approach incorporated in a BOKINETICS code developed at our Laboratory (Garcia et al., 2006), plus a set of Uranium transfer rate parameters as function of individuals' age, assuming an uninterrupted exposure over a period of 60 years.

The concentrations of U in kidneys, liver and blood were calculated for U ingestion rates ranging from 2 to 12mg-U/day, which determined the limits of the data bands (Figure 1). These ingestion rates were inferred from U concentrations measured in teeth (Prado et al., 2008). Results for kidneys and liver are very high, but our calculations revealed an aggravating circumstance in the sense that Uranium concentrations in these organs saturate at ages of 10 and 20 years, respectively. This is of great concern because in this case they would constitute severe radiological burdens inflicted to children and youngsters. Such a circumstance could lead to serious health problems as neoplasias. As a byproduct, this work could subsidize further studies

toward the identification of the driving factors in environmental pollution, associated with possible risks to the public health.

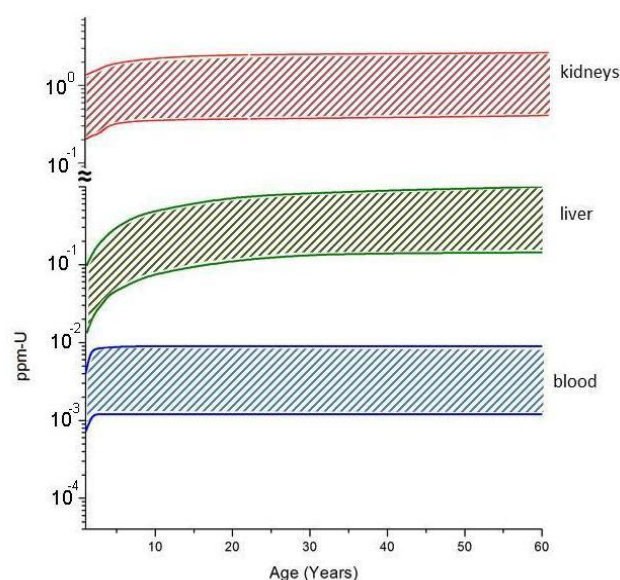


Figure 1. Concentrations of U in kidneys, liver and blood as function of age (see text for details).

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# Radium concentration in (TE)NORM materials by radon emanation prior and after acidic dissolution of the samples

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Keywords: (TE)NORM, acidic dissolution, radium analysis, radon emanation.

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Radium content and radon emanation of environmental samples and building materials such as phosphogypsum and granite (Efsthathiou *et al* (2013)), as well as oil and gas production wastes (Moatar *et al* (2010), Devecchi *et al* (2014)), is of fundamental importance regarding radiological impact assessments and development and application of radiation protection measures. Previous studies have shown that the radon emanation rate of solid samples strongly depends on material properties such as particle size, porosity, humidity etc. (Sakoda *et al* (2011)). In contrast, radon emanation from homogeneous aqueous solutions could be an alternative method to apply radon emanation techniques successfully to a wide range of solid (TE)NORM samples.

The objective of this work was to develop and assess a method for the determination of radium (basically  $^{226}\text{Ra}$ ) in phosphogypsum samples prior and after acidic dissolution by means of a closed recirculating air loop radon emanation system including a radon monitor and a tightly closed aluminium vessel.

The system has been calibrated with radium-containing standard solutions and the data evaluation included linearity, detection limits and analytical resolution of the method. To validate the method described here, phosphogypsum samples of known radium activity determined by gamma spectrometry (GS), have been used. To ensure a constant emanation factor corresponding phosphogypsum samples of 0.7 g were dissolved in 30 ml of 8 M  $\text{HNO}_3$  and during measurement the vessel was thermostated at 23 °C. The radon concentration reaches a maximum value in all cases after 5 days measuring time and the radon ingrowth data are well fitted with the theoretical curve calculated using the radon ingrowth equation.

According to the experimental data (Figure 1) the radon emanation system presents significantly better linear response to the radium content of the dissolved ( $R^2 = 0.94$ ) compared to the powdered samples ( $R^2 = 0.63$ ), the detection limit for radium after acidic dissolution is  $\sim 90 \text{ mBq g}^{-1}$  and the method presents fairly good resolution (100 mBq).

In addition, radon emanation measurements have been performed with granite and uranium mineral samples prior and after acidic sample dissolution. The corresponding data indicate clearly that after dissolution the sensitivity of the method increases significantly, because of the dramatic change in the radon emanation factor.

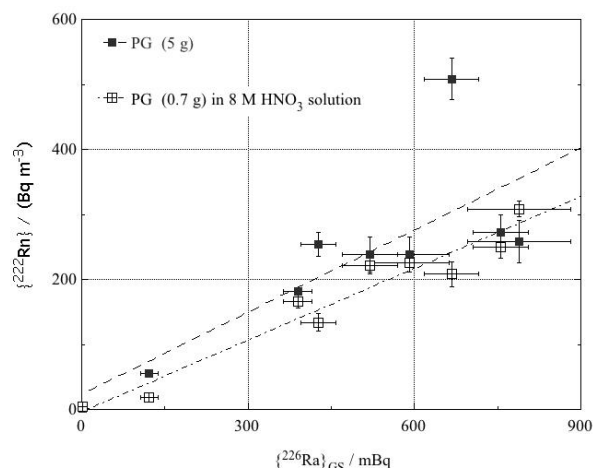


Figure 1. Radon activity concentration ( $\text{Bq m}^{-3}$ ) as a function of the radium content of phosphogypsum samples, determined by the radon emanation system described here, prior and after acidic sample dissolution.

The radon emanation system described here has been effectively applied to radium determination in (TE)NORM samples indicating that the method could be attractive because of its simplicity, robustness, effectiveness and repeatability regarding the radium determination in radium-containing samples by radon emanation measurements.

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# Development and validation of a methodology for determination of U and Th in soil using alpha spectrometry

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Keywords: method validation, uranium, thorium, alpha spectrometry.  
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Among various methods to determine the radioactivity concentration in an environmental sample, alpha spectrometry is a powerful analytical tool for the identification and assay of the alpha-emitting sources primarily due to its high counting efficiency, high sensitivity and low price (Vajda, 2012). To perform the determination of the radioactivity in samples, a sample dissolution should be carried out. In the dissolution methods, a fusion method is a very effective way of rapid and complete sample decomposition. However, large amounts of salts together with impurities are often transferred to the sample solution, which may create problems in the chemical separation.

The objective of this study is to develop a methodology for the determination of U and Th in soil using a fusion technique and alpha spectrometry, and evaluate the methodology by the validation parameters of linearity, selectivity, accuracy, precision, etc.

The overall process from the sample preparation to the quantitative analysis using an alpha spectrometer is presented in Figure 1.

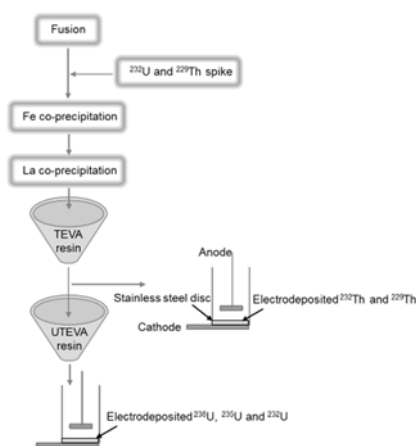


Figure 1. Schematic diagram of procedure for determination of uranium and thorium isotopes in soil.

In brief, a soil sample (NIST, SRM 2709a) was destructed by a fusion technique after ashing the sample. And then  $^{232}\text{U}$  and  $^{229}\text{Th}$  tracer solutions were added into the fused sample. Fe and La co-precipitation procedures were carried out to extract the U and Th in the fused sample. The pre-concentrated sample was passed through the TEVA and UTEVA column in sequence to

obtain pure Th and U isotopes respectively. The obtained isotopes were electrodeposited on a stainless steel disks. And alpha spectrometry measurement was conducted to determine their radioactivity concentration.

The radioactivity of  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  were measured and evaluated for the methodology validation. The linearity on the sample amount vs. activity concentration was evaluated with soil samples, which was prepared variously by inserting the standard solution containing uranium and thorium into the soil sample. Within the range of ca. 40 – 4000 Bq/kg, a very good correlation ( $r^2 = 0.99$ ) was observed for  $^{238}\text{U}$  (Figure 2). Good correlations for other nuclides ( $^{235}\text{U}$  and  $^{232}\text{Th}$ ) were also observed.

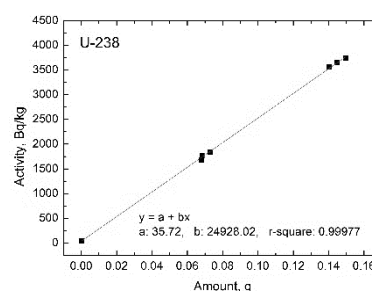


Figure 2. Plot of amount of standard solution in sample vs. activity.

Other validation parameters were also in agreement to use our methodology for the determination of U and Th radioactivity concentration in the soil sample using alpha spectrometry. However, when our methodology was applied to determine the activity in zirconium and bauxite ores, there were large standard deviation due to remained zirconium and aluminium in the electrodeposited layer.

In summary, the developed methodology is very useful to determine the radioactivity of the environmental sample such as soil or rock in a wide range of activity concentration, and offers good prospects for its application in routine monitoring programmes.

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# Method Validation for the Quantification of Natural Radionuclides Using a Gamma-Ray Spectrometry in Raw Materials and By-products

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Keywords: method validation, natural radionuclides, gamma-ray, raw materials, by-products.  
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According to the “Act on protective action guidelines against radiation in the natural environment” in Korea, the methodology for an analytical evaluation on natural radionuclides was designed and developed, focusing especially on the raw materials and by-products in domestic distribution. Since the analysis method for natural radionuclides demands a systematic procedure and QA program, it is necessary to conduct a method validation for the analysis procedures developed.

A gamma-ray spectrometry still has an effect on the assessment of the radioactive concentration for natural radionuclides with a direct measurement, such as  $^{40}\text{K}$  as well as an indirect measurement using its progeny as an indicator, such as  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  (Ji, 2015). Table 1 shows the natural radionuclides to be analysed with direct and indirect measurement using a gamma-ray spectrometry. In the table, lab-scaled recovery means that the radioactive equilibrium between  $^{226}\text{Ra}$  and  $^{214}\text{Bi}$  or  $^{214}\text{Pb}$  should be attained by the confinement of  $^{222}\text{Rn}$ , which is a noble gas, in a sample container.

Table 1. Natural radionuclides to be analysed.

	Targent nuclide	Indicator	Remarks
Direct	$^{40}\text{K}$	-	-
	$^{238}\text{U}$	$^{234}\text{Th}$ , $^{234\text{m}}\text{Pa}$	-
Indirect	$^{226}\text{Ra}$	$^{214}\text{Bi}$ , $^{214}\text{Pb}$	Lab-scaled recovery
	$^{232}\text{Th}$	$^{228}\text{Ac}$	-

In this study, a method validation for the analysis procedure of natural radionuclides with a gamma-ray spectrometry was conducted according to the ISO/IEC 17025, which demands performance tests, such as accuracy including trueness and precision, linearity, selectivity, repeatability or reproducibility, limit of detection (LOD) and quantification (LOQ) and robustness. First, the representative sample matrix to be used in the validation was selected to be zircon and bauxite series as raw materials, and ash and dust in treating them as by-products. The acceptance criteria for the defined validation parameters, such as the linearity, trueness, and precision, were then established according to the IAEA method, which means the relative bias, U-test, and P-value (IAEA, 2011).

Standard samples within an Al can with a volume of about 100 mL to confine radon gas were prepared with six radioactive levels of natural radionuclides from about 50 to 1,500 Bq/kg. After attaining the secular equilibrium

during about 4 weeks, the replicate was conducted according to the validation protocols developed.

First, the linearity was determined from the 3 replicates of six standard samples, as shown in Figure 1, and below 10 % of the relative bias was maintained in all replicates. This means the reliable results of the analytical assessment of natural radionuclides using gamma-ray spectrometry can be confirmed within an activity range of about 50 to 1,500 Bq/kg.

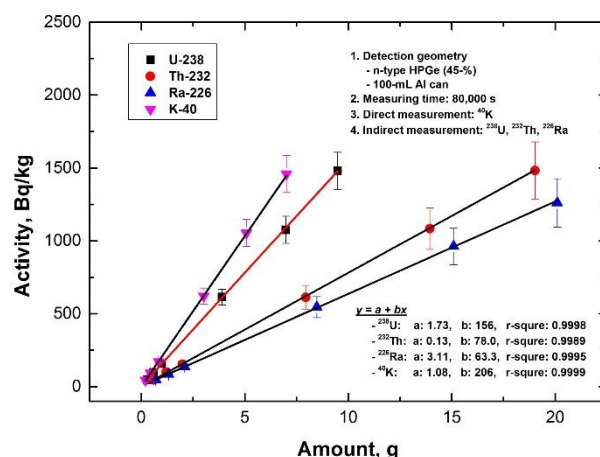


Figure 1. The linear range in the gamma-ray spectrometry for natural radionuclides.

The accuracy from 7 replicates was evaluated at three different activity levels, i.e., below 100 Bq/kg, from 100 to 1,000 Bq/kg, and above 1,000 Bq/kg and could reach the excellent results of below 1.0 in U-value and 15 in P-value for the trueness and precision. The LOD of natural radionuclides was determined to be about 25, 20, 5, and 5 Bq/kg for  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{226}\text{Ra}$ , respectively, in the case of using a HPGe detector with a 40 % relative efficiency during a measurement of 80,000 seconds. The robustness was also evaluated from a NIST SRM 1633c and 600, which means coal fly ash and bauxite. Good results were achieved at the relative bias and relative standard deviation for the replicates.

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# Effective dose assessment due to Águas de Lindóia water ingestion

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The radionuclides from U and Th series are responsible for about 80% of the natural radiation exposure, together with the <sup>40</sup>K. These primordial radionuclides are widely spread in the lithosphere, atmosphere and hydrosphere. The main route for its internal exposition is through ingestion. In the water, the radionuclides can be dissolved, as complexes, or bound to the particulate matter.

The consumption of mineral water with relatively high radioactive levels is a generalized practice for medicinal purposes. Águas de Lindóia located in São Paulo State, Southwest, Brazil, is a well known region where radioactive water is found and several spas offer therapeutic, cosmetic and anti-stress treatment.

The Águas de Lindóia waters had already been characterized for the long lived radio isotopes (Negrão, 2012), but data on a complete evaluation of the effective dose assessment are scarce. The objective of this study was to evaluate de effective dose due to ingestion of Águas de Lindóia waters due to the presence of <sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>210</sup>Pb and <sup>40</sup>K.

Neutron activation analysis were applied for the <sup>238</sup>U, <sup>232</sup>Th and K determination while total alpha and beta counting scintillation were use to the determination of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>210</sup>Pb.

For neutron activation analysis determinations, 2 L of the water sample was evaporated to 20 mL. From this concentrated sample, 1mL was taken and transferred to a paper filter sheet and dried under an infrared lamp. Synthetic standards were also prepared by pipetting convenient aliquots of standard solutions (SPEX Certiprep Inc., USA) onto small filter paper sheets. The samples and synthetic standards were irradiated for 8 h, in the IEA-R1 nuclear reactor at IPEN under a thermal neutron flux of  $1 - 5 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ . The counting was done at different time frames, five days after irradiation for U and K and 20 days after for Th determination, by Gamma Spectrometry using an EG&G Ortec Ge Hyperpure Gamma Spectrometer detector (AMETEK Inc., USA) and associated electronics, with a resolution of 0.88 keV and 1.90 keV for <sup>57</sup>Co and <sup>60</sup>Co, respectively. The analysis of the data was done by using an in-house gamma ray software, VISPECT program, to identify the gamma-ray peaks.

For <sup>228</sup>Ra, <sup>228</sup>Ra and <sup>210</sup>Pb determination the following procedure was adopted. Carriers of Ba<sup>2+</sup> and

Pb<sup>2+</sup> were added to 2 L of water samples. The solutions were treated with citric acid for iron and lead complexation. Sulfuric acid was added for Ra<sup>2+</sup> sulfate precipitation, co-precipitation as Ba(Ra)SO<sub>4</sub> and PbSO<sub>4</sub>. The precipitate was dissolved with NTA and 6M NaOH was added. Solutions of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and glacial acetic acid were added to precipitate Ba(Ra)SO<sub>4</sub> leaving Pb<sub>2+</sub> in solution. The precipitate was separated in two steps of centrifugation, dissolved with EDTA and again precipitated as Ba(Ra)SO<sub>4</sub>, that was filtered in Millipore filter and stored for counting. Counting was performed after 21 days of precipitation. The solution containing Pb<sub>2+</sub> was treated with 1M Na<sub>2</sub>S<sub>2</sub> to precipitate PbS. The precipitate was centrifuged, dissolved in HNO<sub>3</sub> and filtered to separate the precipitated sulfur. The addition of 30% Na<sub>2</sub>CrO<sub>4</sub> precipitated PbCrO<sub>4</sub> that was filtered in Millipore filter and stored for counting. Counting was performed after 10 days of precipitation. Counts were made in a gas flow proportional detector of low background, Berthold, model Lb 770, during 200 minutes. The procedure was taken from Moreira (1993) and Oliveira (1993).

Results showed that the moderate consumption of the Águas de Lindóia water is unlikely to exceed the annual permitted limit of 1 mSv/y.

## Acknowledgments

This work was supported by the National Council for Environmental Radioactivity under grant AI/2014.

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# Integrated doses from natural radioactivity measurements by using LiF:Mg,Cu,P and Al<sub>2</sub>O<sub>3</sub>:C thermoluminescence dosimeters (TLD).

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Keywords: thermoluminescence, dosimetry, radioactivity, TLD.  
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**Introduction:** Luminescence has been well established as an effective as well as a versatile dosimetric technique in various fields such as medical, environmental, personal, space and retrospective dosimetry, (Aitken, 1985; Bailiff, 1994). Luminescence techniques are frequently applied in archaeological science for absolute dating studies (Aitken, 1985) or for characterization of materials of archaeological interest (Liritzis, 1997).

For an artifact which has been heated, the TL signal is proportional to the yearly rate at which new TL is created multiplied by the number of years since the sample was last heated. New TL results mainly from the decay of natural radionuclides, such as <sup>232</sup>Th, <sup>40</sup>K and natural U, along with cosmic rays, which provide a constant source of low-level ionizing radiation. The accumulated dose during the past is termed as paleodose or equivalent dose. Furthermore, the rate at which this energy-dose is accumulated is termed as dose rate.

The annual dose (dose-rate) is estimated with natural radioisotopes, such as <sup>40</sup>K, <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th.

The interest of the present study is to find out a new method in order to measure the integrated dose which is released due to the presence of natural radioactive elements such as <sup>40</sup>K, <sup>238</sup>U, <sup>226</sup>Ra, <sup>227</sup>Ra and <sup>232</sup>Th into several geological samples. The main impact of these results is that through this method is possible to evaluate the annual dose.

The geological samples used in present study have already been measured for their natural radioactivity using  $\gamma$ -spectrometry methodology (Papadopoulos et al., 2013)

**Apparatus and measurement conditions:** TL measurements were carried out using an automated Risø TL/OSL reader, model TL/OSL-DA-15, equipped with an internal <sup>90</sup>Sr/<sup>90</sup>Y beta source (~0.1 Gy/s). Luminescence signals were detected using an EMI 9635QA photomultiplier using a combination of a Pilkington HA3 heat absorbing and a Corning 7–59 (320–440 nm) blue filter. A heating rate of 1°C/s was used in all TL readouts in order to avoid significant temperature lag, up to a maximum temperature of 400°C.

**Experimental:** The experiment was divided into two different experimental procedures:

- For the first procedure were selected, from the geological sample, the coarse grains from 40µm to 80µm. These grains were mixed with TLD's grains, sized more than 80µm. The TLD used for this situation is a widely used dosimeter, the lithium fluoride (LiF:Mg Cu,P). After the end of exposure time the TLD gets separated from the sample simply by sieving.

- For the second procedure were selected grains less than 40µm from the geological sample and were mixed with TLD's grains between 2-10µm. The TLD used in this procedure is aluminum oxide (Al<sub>2</sub>O<sub>3</sub>:C). Grains between 2-10µm were collected by applying the fine grain method (Zimmerman, 1971). The sample was crushed gently in an agate pestle and mortar. The fine grains between 2-10µm were separated from coarse grains by sinking them into acetone solvent. In that case the dosimetric material is separated by geological material by applying once more the fine grain method.

Discs made of aluminum substrate 0.5 mm thick and 9 mm in diameter, each carrying 1-2mg of the sample, were prepared. The preparation and analysis of samples was formed under the red light illumination.

Specific activities of <sup>40</sup>K, <sup>226</sup>Ra and <sup>232</sup>Th (Bq·kg<sup>-1</sup>) and the lab-names of geological materials used are presented in Table 1.

Table 1: Specific activities of <sup>40</sup>K, <sup>226</sup>Ra and <sup>232</sup>Th (Bq·kg<sup>-1</sup>). (Papadopoulos et al., 2013)

Origin	Name	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Xanthi	GAE1	15.6	18.2	291
	GAE11	5.3	6.5	175
Papikio	P220	57.9	69.4	1173
Kavala	KB6	192.3	106.4	754.4
Flamouri	FL1	29.9	12.4	484.4
Monopigado	MO4	34.9	41	758
Fanos	MD2	76.2	209	1245
Kastania	KST20	31.9	52.4	918.7
	KST5	14.8	44.1	707.7
Naxos	VN1	99.8	131.4	1140.7
Ikaria	PI1	183.3	63.5	1144.7

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# Radon (Rn-222) determination in Portuguese waters using two phase liquid scintillation counting (LSC) technique

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The radon (Rn-222) is a naturally occurring gas, from radioactive uranium series, which is formed by radioactive decay of Ra-226 its parent. This radionuclide is found disseminated throughout the earth's crust in varying amounts depending on the composition of geological substrate. The Rn-222 is dissolved in water and could represent a public health hazard by ingestion or by inhalation after emanation of water (WHO, 2009). The importance of the detection of radon in water is associated with the fact that some ground waters, from granite formations, may contain high concentrations of natural radionuclide of uranium series, leading to high levels of Rn-222.

In this paper, we describe the method implemented for the determination of Rn-222 dissolved in water by liquid scintillation counting (LSC) technique. After sampling, a volume of 10 mL of sample was withdrawn with the aid of a syringe and injected slowly below 10 mL of the specific scintillation cocktail surface, immiscible with water, without causing turbulence in order to minimize any loss of radon (Lopes *et al*, 2014). Before measurement, the sample is stored for 3 hours until equilibrium is reached between Rn-222 and its alpha emitting decay products. The alpha activity is measured in a liquid scintillation counter (Tri-Carb 3170 TR/ SL) during 60 minutes of counting time, using alpha counts only, through alpha-beta discrimination. For calibration of the detection system, standards of Ra-226 were prepared and measured after a period of about 30 days, for Rn-222 growth.

The quality control of the Rn-222 measurements was assessed by evaluation the precision and accuracy of the two phase LSC method, based on repeatability testing and interlaboratory comparison (ILC) exercises. The results submitted in the framework of radon in drinking water ILC exercise are in good agreement.

Several water samples were analyzed from different underground captures. About 50% of the total samples have Rn-222 concentrations  $<100$  Bq L<sup>-1</sup>. In general, the results show that these waters have acceptable levels of Rn-222 in accordance of European recommendation (EURATOM, 2001) However, waters originating from granite areas formations have shown higher activities (e.g.  $\approx 1300$  Bq L<sup>-1</sup>).

National waters should keep monitored in order to determine their radioactivity content, especially in areas where it is suspected that the concentrations are higher than the parametric value set to radon in water intended for human consumption (EURATOM, 2013).

The methodology applied seems to be appropriate and the successful external evaluation of Rn-222 measurements allows now the possibility to submit the method to an accreditation.

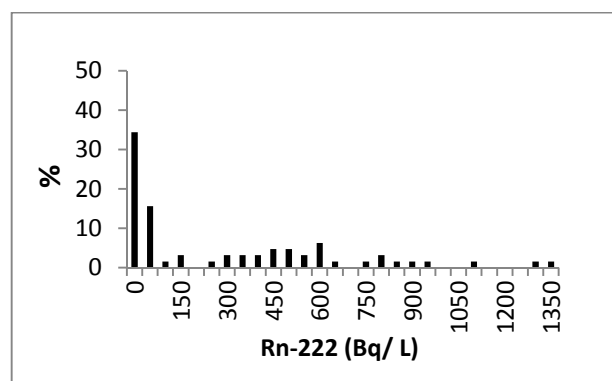


Figure 1. Rn-222 activity in natural ground waters

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# Indoor radon exposure in some energy- efficient houses from Romania

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Reconstruction techniques such as the thermal insulation or enhancing air exchange or using fans could have a negative impact on energy consumption (Pressyanov et al, 2015). Modern trends in civil construction are based on increasing the energy efficiency of buildings. In the light of the ongoing policy to improve the energy efficiency of existing buildings, it is essential to evaluate the effect of new construction methods on the indoor radon level.

In the present paper the exposure to radon in some energy efficient buildings from Romania was discussed. To investigate the influence of modern buildings technologies on the accumulation of radon in the indoor atmosphere, detailed measurements of radon will be performed in 25 energy efficient houses constructed in the last decade by using mostly solid concrete or gas-concrete blocks. Indoor radon measurements were performed by using nuclear track detectors CR-39 exposed for 6-9 months on ground floor levels of dwellings, according to the NRPB Measurements Protocol.

The exposure of indoor radon in the 53 studied rooms from energy efficient houses involved in study varied from 19 to 792 Bq m<sup>-3</sup> with a mean of about 168 Bq m<sup>-3</sup>. With respect to the type of surveyed rooms within dwellings, the descriptive statistics of measurements by room type is summarized in Table 1.

Table 1. Descriptive statistics for the radon measurements by room type within dwellings of Romania.

Room	Frequency	AM $\pm$ SD (Bq m <sup>-3</sup> )	Max. (Bq m <sup>-3</sup> )	% (No.) > 300 (Bq m <sup>-3</sup> )
Living	26	160 $\pm$ 123	792	15(4)
Bedrooms	23	189 $\pm$ 179	667	22(5)
Kitchen	4	99 $\pm$ 70	171	0
Total	53	168 $\pm$ 147	792	17(9)

The AM indicate that the highest radon concentrations occur in bedrooms, although in particular the highest value of radon concentration was found in a living room (792 Bq m<sup>-3</sup>). The variability of radon concentrations among surveyed houses and within rooms could be related to a series of factors, such as the

type of building (number of rooms at ground floor), type of thermal insulation, the behavior of the occupants, various air exchange systems and age of construction (Gräser et al, 2010).

The obtained value is approx. 33% higher than the average reported by authors for conventional homes of Transylvania, Romania (Cosma et al, 2013).

With respect of the year of construction, the mean values of radon concentration in the group of buildings constructed in the last decade are well greater than the average indoor radon level in buildings constructed during 1940-1990, which confirms the assumption that new energy efficient technologies or rehabilitation techniques effects on radon accumulation levels in dwellings.

The high levels of radon concentration related to the energy efficient reconstruction can be reduced by ventilation. Although it is one of the possible techniques, which effectiveness is amply studied, it is advisable to put emphasis on the energy impact on loss of thermal comfort conditions that implies.

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# Application of the ERICA Assessment Tool for the calculation of the dose rates in terrestrial herbivore mammals in Greece

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In this study the ERICA Assessment Tool (Version 1.2.0, November 2014) (Brown et al., 2008) was applied for the calculation of the dose rates received by terrestrial herbivore mammals due to the exposure to natural and artificial gamma emitting radionuclides.

Samples of soil and herbivore mammals were collected from semi-natural grassland areas in Greece, covered with typical vegetation of the Eastern-Mediterranean ecosystem. The collected animals were herbivore mammals (sheep and goats of the Bovidae family) of free-range grazing, intended for human consumption (food and skin products) (Sotiropoulou et al., 2013). The organisms and the abiotic samples were measured for natural and artificial gamma emitting radionuclides (<sup>226</sup>Ra, <sup>228</sup>Ra, <sup>228</sup>Th, <sup>137</sup>Cs, <sup>134</sup>Cs, <sup>131</sup>I) by the use of gamma spectrometry. The detected artificial radionuclides are residuals from the global and the Chernobyl fallout, and traces of the latest accident in Fukushima as well (Kritidis et al., 2012; Sotiropoulou et al., 2012).

For the simulation of the transfer of the studied radionuclides and the dose rates calculation a simple food web was considered, consisting of the representative organisms of the specific ecosystem type. For this purpose a phantom organism was created through the ERICA Tool based on the ecosystem features and the biometric characteristics of the studied organisms. Besides the measured parameters, the simulations were conducted taking also into

consideration the appropriate default radiological and ecological parameters of the datasets of the Tool. The calculations of the dose rates were performed on the basis of internal dose rates from the activity concentrations in mammals, external dose rates from the activity concentration in soil and total dose rates from the combination of the previous two.

The calculated dose rates of the selected organisms were below the default screening level of 10  $\mu\text{G h}^{-1}$ . Even though the ERICA Tool is mainly a generic model, when real-time measurements are introduced, can result to more specific findings in terms of the ecosystem considered. Thus, it is a useful model that can be applied for the radiological impact assessment on the basis of either simulations or predictions.

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# External and internal annual effective doses for Paraná state granites used as internal coating building materials by high resolution gamma-ray spectrometry

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Keywords: effective dose, granites, radon, gamma spectrometry, building materials  
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The entire terrestrial crust, including rocks and soils, contains different amounts of natural radioactive nuclides, mainly primordial radionuclides as the single radioactive isotope of potassium  $^{40}\text{K}$  and radionuclides from the uranium  $^{238}\text{U}$  and  $^{232}\text{Th}$  series. As a consequence, geological materials used as building materials act as a source of radiation and in massive houses made of brick, concrete or stone the absorbed dose rate depends mainly of the activity concentration of natural radionuclides in those building materials.

A complete evaluation of the annual effective dose in a construction should consider both the external and internal contributions of the constituent materials.

In this work, the potential radiological hazard for 37 commercially-used granites, used as coating building materials, extracted in outcrops from the crystalline basement of Paraná state Brazil, mainly of Curitiba Metropolitan Region, was assessed through the  $^{226}\text{Ra}$  ( $^{238}\text{U}$  series),  $^{232}\text{Th}$  and  $^{40}\text{K}$  activities concentrations, determined by high-resolution gamma-ray spectrometry.

The external contribution was evaluated by using the  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  activities concentrations in all employed materials, weighted by their fraction in the construction and also considering the background radiation of the studied location.

The internal contribution due to indoor radon inhalation was evaluated through the  $^{226}\text{Ra}$  activities concentrations.

In order to determine the annual effective dose, the radon exhalation rate was determined by a simplified mathematical equation for construction materials (UNSCEAR, 2000).

The increment of dose by external gamma rays and the internal dose due radon inhalation were simulated as suggested by the European Commission of Radiological Protection (EC, 1999) for a model room with all internal walls of dimensions 4 m x 5 m x 2.8 m coated with the studied rocks having 3 cm thickness and 2600 kg.m<sup>-3</sup> density, for an annual exposure time of 7000 hours, a dose conversion factor of 0.7 Sv.Gy<sup>-1</sup> and a background radiation of 50 nGy.h<sup>-1</sup>.

The results for external and internal annual effective dose estimated from the absorbed dose rate radon exhalation rate, are shown in Fig. 1. The estimated values from the contribution of inhaled radon, varied from  $(21 \pm 2) \mu\text{Sv.y}^{-1}$  up to  $(432 \pm 14) \mu\text{Sv.y}^{-1}$ . The total annual effective dose, considering the sum of these two contributions, ranged from  $(42 \pm 2) \mu\text{Sv.y}^{-1}$  up to

$(655 \pm 15) \mu\text{Sv.y}^{-1}$ , below the recommended EC value of 1 mSv.y<sup>-1</sup> (EC, 1999).

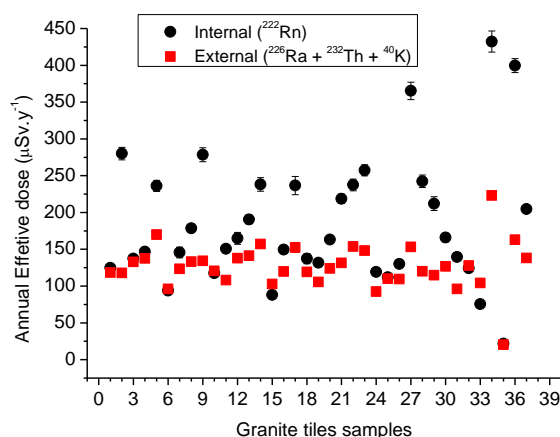


Figure 1. Internal and external annual effective doses for Paraná granites used as coating building materials.

The calculated values are below the ICRP 60 recommended limit for general public of 1 mSv.y<sup>-1</sup>, but one should keep in mind that only the dose increment due to coating materials in the described scenarios was evaluated, and, for a complete assessment, the contribution of all construction materials must be considered.

## Acknowledgments

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# Absorbed dose estimation to family members of patients treated with radioiodine for thyroid cancer

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Keywords: absorbed dose, thyroid cancer, radioiodine, family members.

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**Background.** Thyroid remnant ablation with radioiodine is a well established treatment for patients with Differentiated Thyroid Carcinoma (DTC) after thyroidectomy. After 3-4 days of hospitalization, these patients return to their homes, presenting a possible radiation hazard to their family members.

**Objectives.** The present work aims at estimate the radiation burden to the cohabitants of DTC patients treated with radioiodine, after their release from the hospital.

**Methods.** 175 patients 48 men, 127 women) suffering from DTC were treated with radioiodine activities ranging from 1850 to 9250 MBq after thyroidectomy. All patients were kept in isolation for 3-4 days into the dedicated shielded room in the Nuclear Medicine Department. Dose rate measurements at a distance of 1 and 2 meters from the patients were performed at regular time intervals throughout their hospitalization. These measurements were used to calculate the effective half life ( $T_{eff}$ ) of radioiodine for each patient. All patients were released from the hospital after the 3rd day, provided that the dose rate at 1 meter ( $D_{out}$ ) had dropped below 10  $\mu\text{Sv/h}$ . In case it hadn't, they were kept in isolation for one more day, or they were released with strict instructions to avoid contact with other people for a few more days. Oral and written instructions with the necessary precautions concerning the minimization of exposure to family members and members of the public were given to all patients, according to their specific family and social status.

Patients were divided into groups according to the age of their cohabitants: adults, children (5-18 years old) and small children (0-5 years old). Different contact times with cohabitants were assumed in each case, in order to estimate the total radiation dose ( $D_{\infty}$ ) to each family member according to the following formula:

$$D_{\infty} = 1.44 D_{out} T_{eff} E / r^2$$

where  $r$  is the distance from the patient and  $E$  is the occupancy factor at that distance. If a patient lived only with other adults, it was assumed that  $E=1/10$  at  $r=1\text{m}$ , provided that the instructed precautions were followed. If children and/or small children were also present,  $E$  was taken equal to  $1/4$  at  $r=1\text{m}$ . If the patient lived with children and/or small children and there were no other adults present,  $E$  was taken equal to  $1/2$  at  $r=1\text{m}$ .

Patients living more than 30 km away from the hospital returned home by car or taxi. The duration of each journey ( $t_j$ ) was recorded and the absorbed dose to the co-traveller ( $D_{tr}$ ) was estimated considering a distance of  $r=1\text{m}$  throughout the journey.

**Results.** Mean Radioiodine  $T_{eff}$  was 15.3 hrs (range 1.9 - 37.5). 137/175 patients (78.3%) were released from the hospital 72 hours after radioiodine administration. The remaining 38/175 (21.7%) had to stay for one more day until the dose rate at 1m had dropped below 10  $\mu\text{Sv/h}$ .

20/175 patients (11.4%) lived alone. 102/175 (58.3%) patients lived together with adults only. 48/175 (27.4%) patients lived with at least one child and/or at least one small child, but they all had at least another adult to take care of the children. Only 5/175 (2.9%) patients lived with one or more children that they had to care for without the help of any other adult. Table 1 summarizes the total absorbed dose to cohabitants and co-travellers, as calculated for each case.

Table 1. Total absorbed dose ( $D_{\infty}$ ) to family members.

	Mean ( $\mu\text{Sv}$ )	Range ( $\mu\text{Sv}$ )
adults	19.5	0.6 - 122.1
children	47.7	4.0 - 540.0
small children		
children	18.5	4.0 - 45.8
co-travellers	12.3	0.2 - 186.7

**Conclusion.** Mean absorbed doses to all family members were estimated to be generally low, well below the corresponding dose limits and dose constraints, mainly due to the fast clearance of radioiodine from patients' bodies. The maximum dose to any child was estimated to be slightly above 0.5 mSv, almost half the corresponding dose limit (1 mSv). The maximum dose to any non-relative co-traveller was less than 0.2 mSv, which is less than the corresponding dose constraint (0.3 mSv).

It can be concluded that, provided that the necessary precautions are followed, the radiation burden to the family members of DTC patients treated with radioiodine after thyroidectomy can be kept much lower than the corresponding dose limits.

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# Natural Radioactivity Levels in the Shallow-sea Hydrothermal System off SE Milos Island, Greece

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**Keywords:** natural radioactivity, marine sediment cores, hydrothermal system, Milos.

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Milos volcanic Island, part of the Hellenic Volcanic Arc (HVA) in Aegean Sea, exhibits a unique shallow-sea hydrothermal system at the prolongation of Fyriplaka volcano. The most intense submarine venting is manifested offshore SE Milos by the colored hydrothermal precipitates on the seafloor (Fig. 1), formed as the hydrothermal fluids discharge through the sediments.

Previous studies concern scattered measurements of the natural radioactivity around the island with respect to soil, ores, marine sediments and seawater (Florou *et al.*, 2007 and references therein). The present work aims to investigate, for the first time, the levels of natural radioactivity ( $^{238}\text{U}$ ,  $^{232}\text{Th}$  decay series and  $^{40}\text{K}$ ), as well as manmade  $^{137}\text{Cs}$  in marine sediment cores from the active hydrothermal system off SE Milos.

Samples were collected by SCUBA diving from: a) the white hydrothermal precipitates, collected in slices of 2 cm down to 22 cm depth, b) the seagrass area, where sampling reached the depth of 12 cm and c) the brown-grey area that is considered the background sediment, down to 20 cm depth.

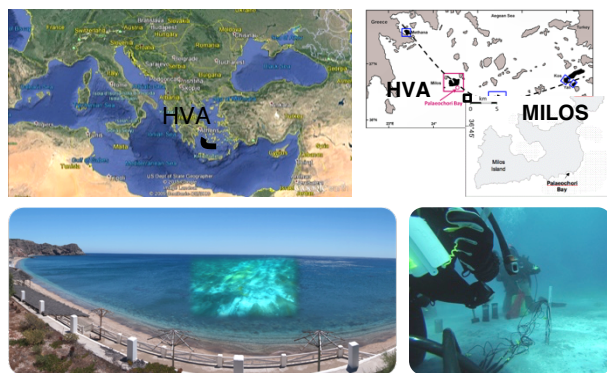


Figure 1. SE Milos shallow-sea hydrothermal system.

The samples have been analyzed with the UoA GEROS  $\gamma$ -ray spectrometry station (a 23% intrinsic efficiency HPGe detector) and the spectra with the SpectrW software suite (Kalfas, 2015). Depth profiles of the activities (in Bq/kg) for  $^{226}\text{Ra}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  originating from  $^{238}\text{U}$  decay,  $^{208}\text{Tl}$  and  $^{228}\text{Ac}$  deriving from  $^{232}\text{Th}$  decay,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  have been prepared and analyzed.

Generally, the depth profiles follow the same trend; the highest values are recorded near the surface and between 14-16 cm depth, whereas the lowest in 12 cm and 20 cm depth. The average, minimum and maximum measured values from the white-area marine core are presented in Table 1. The artificial radionuclide,  $^{137}\text{Cs}$ , was not observed in significant amounts above the background.

Table 1. Average, minimum and maximum values in Bq/kg for the white area, 0-22 cm depth.

Radionuclide	Average value Bq/kg $\pm$ error	min-max
$^{226}\text{Ra}$	$603 \pm 4$	378-898
$^{214}\text{Pb}$	$18.3 \pm 0.2$	15-23
$^{214}\text{Bi}$	$24.9 \pm 0.3$	21-32
$^{228}\text{Ac}$	$18.6 \pm 0.4$	15-23
$^{208}\text{Tl}$	$55.4 \pm 1.8$	25-67
$^{40}\text{K}$	$530 \pm 6$	491-650
$^{137}\text{Cs}$	$1.42 \pm 0.03$	0.6-2

Average values are in agreement with previous work in Milos and comparable to other hydrothermal settings from Greece. However, this is not the case for  $^{226}\text{Ra}$ , which is found to be higher than previously reported.

## Acknowledgments

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# Development of rice reference material and a proficiency test for the measurement of Cs-137 and K-40

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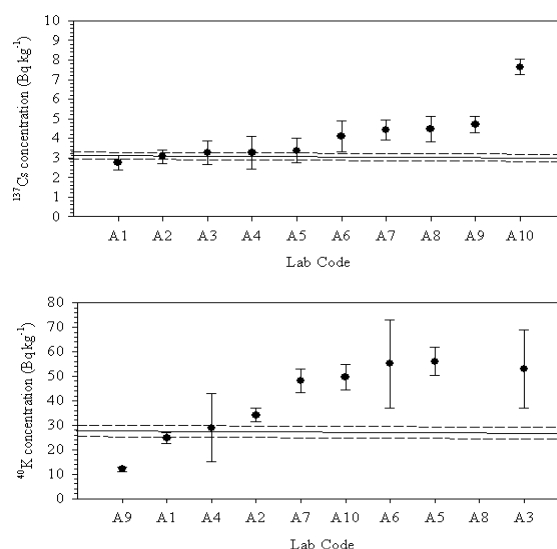
Keywords: Reference Material, Rice,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ . Intercomparison exercise  
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For a laboratory to produce consistently reliable data, it must implement an appropriate programme of quality-assurance and performance-monitoring procedures. Intercomparison exercise is one of these procedures. The usual format for Intercomparison exercise schemes is based on the distribution of samples of a test material to the participants. In accordance with the RCA (Regional Cooperative Agreement) /UNDP(United Nation Development programme) Project Work Plan, the international intercomparison exercise was organized and 10 Asian countries participated on the programme. The purposes of the intercomparison exercise were to provide the quality assurance on the data produced by the participating laboratories, to review performance-monitoring procedures and to exchange technical information.

The reference material used for the intercomparison exercise was rice material which was developed by KRISS for the measurement of Cs-137 and K-40.

To produce rice bearing Cs-137, KRISS was in collaboration with KAERI (Korea Atomic Energy Research Institute, Korea). The rice culture was conducted from the paddy field in the greenhouse located in KAERI. An aqueous carrier-free radioactive solution (74-93 kBq mL<sup>-1</sup> as  $^{137}\text{CsCl}$ ) was applied to distribute uniformly on the water surface and then mixed with the topsoil to about a 15 cm depth (Choi et al., 2005). As the quantity of material obtained from KAERI was small and the activity of Cs-137 was too high to carry out PT, the sample was mixed with natural rice ( $\text{Cs-137} < \text{MDA}$ ).

The homogeneity on the rice material was tested by measuring Cs-137 and K-40. The rice samples were stored in 4 bulk containers, and 2 samples (total 7) were taken from each bulk container and Cs-137 and K-40. The low-level background gamma-ray spectrometer (LB-HPGe) developed by KRISS (the relative efficiency 120 %) was employed to determine the reference activity for Cs-137 and K-40 in the rice sample. The integral background of counting rate of LB-HPGe detector was 1.61 s<sup>-1</sup> from 50 to 3000 keV and the count rate of  $^{40}\text{K}$  measured at 1461 keV was 1.01×10<sup>-3</sup> s<sup>-1</sup>. The relative uncertainties of homogeneity are estimated to be 2.2 % for Cs-137 and 3.5 % for K-40, respectively. The reference values and the participant's results for Cs-137 and K-40 showed at Fig. 1.



While the results of Cs-137 reported by participants were relatively concordant with the reference value, K-40 results were higher than the reference value. This is attributable to the difficulty of K-40 measurement due to the low concentration of K-40 in the rice reference material and a high background by contributing a natural K-40 radioactivity.

## Acknowledgments

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# Method Validation for the Quantification of Radioactivity concentration in NORM samples using ICP-MS

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Keywords: method validation, natural radionuclides, ICP-MS, LiBO<sub>2</sub> fusion.  
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Naturally occurring radioactive materials (NORM) are widely spread throughout the environment. Concern regarding the radioactivity from these materials has therefore been growing over the last decade. To determine the proper handling options, a rapid and accurate analytical method that can be used to evaluate the radioactivity of radionuclides (e.g., <sup>238</sup>U, <sup>235</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra, and <sup>40</sup>K) should be developed and validated.

A measurement technique using ICP-MS allows radioactivity in many samples to be measured in a short time period with a high degree of accuracy and precision. However, the pretreatment process consequently plays an important role in the measurement uncertainty. Thus, a method development and validation should be performed.

In this study, a method was validated according to the ISO/IEC 17025 for a rapid analysis of natural radioactive nuclides (<sup>238</sup>U, <sup>235</sup>U, and <sup>232</sup>Th) using ICP-MS. Method validation parameters (e.g., accuracy, precision, linearity, selectivity, repeatability or reproducibility, limit of detection (LOD) and quantification (LOQ) and robustness) were derived for the various CRM samples.

The method (Fig.1) was established using alkali-fusion with LiBO<sub>2</sub> and ICP-MS measuring technique. A magnetic sector field ICP-MS (SPECTRO MS) was used for a rapid determination of the target radionuclide concentration.

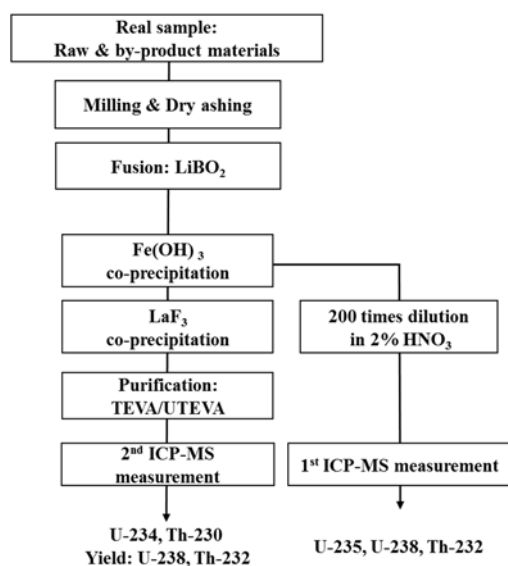


Fig. 1. Analytical procedure using ICP-MS for NORM samples

The LOQ was evaluated using the mean and standard deviation of background counts for the analytical blank samples. According to the established determination method, the LOQ of <sup>232</sup>Th and <sup>238</sup>U were 0.475 Bq/kg and 2.580 Bq/kg, respectively. These values fell below enough minimum detectable range to the regulation guide levels for both nuclides in Korea.

For determining the accuracy and precision of the established method, various CRMs of soil, coal ash, rock, bauxite, and zircon matrix (e.g., NIST SRM 2709a, 1633c, 278, 600, and BCS 388) were analysed. It was found that relative biases of all target nuclides (against CRM values) fell below 10%. Repetitive analyzes (n=7) yielded the relative standard deviations below 5% for all target nuclides. Thus, the analytical results for U and Th derived using LiBO<sub>2</sub> fusion and ICP-MS determination were fairly reliable.

The radioactivity of <sup>238</sup>U, <sup>235</sup>U and <sup>232</sup>Th were analyzed in the raw material samples (N=15) collected from various manufacturing factories in Korea. It was found that the precision of the repetitive measurement results for each sample were less than 10% for all samples.

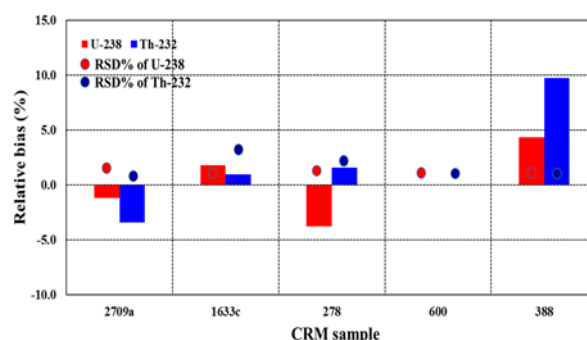


Fig. 2. Accuracy and precision of the analytical results

According to the validation results, we could conclude that the alkali fusion using LiBO<sub>2</sub> flux could be used for rapidly and fully recovering of U and Th elements from the samples and a measurement technique using ICP-MS allow radioactivity in many samples to be measured in a short time period with a high degree of accuracy and precision.

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# On using non-destructive neutron activation analysis to effectively determine $^{235,238}\text{U}$ , $^{40}\text{K}$ and $^{232}\text{Th}$ in various matrices with sub-gram quantities of material

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Keywords: neutron activation analysis,  $^{235,238}\text{U}$ ,  $^{40}\text{K}$ ,  $^{232}\text{Th}$   
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The naturally occurring radionuclides of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  and their daughter products are a potential major source of naturally occurring radiation found in various matrices. We have determined these three radiation sources in geological, food and tobacco. We used the following NAA reactions of  $^{238}\text{U}(n,\gamma)^{239}\text{U}$ ,  $^{232}\text{Th}(n,\gamma)^{233}\text{Th}$  and  $^{40}\text{K}(n,\gamma)^{42}\text{K}$  to determine  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively.

The activity of  $^{238}\text{U}$  can easily be determined by epithermal neutron activation analysis. The use of epithermal neutrons, as opposed to typical thermal NAA, allows for better counting statistics at low uranium concentrations; this is due to both the large epithermal absorption cross-section of  $^{238}\text{U}$  and the low epithermal absorption cross-sections of  $^{37}\text{Cl}$  and  $^{23}\text{Na}$ . The  $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$ ,  $^{37}\text{Cl}(n,\gamma)^{38}\text{Cl}$  and  $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$  reactions typically increase the Compton continuum of irradiated biological samples thus reducing the ability to detect radionuclides at low-level concentrations. Using isotopic ratios, the activity due to  $^{40}\text{K}$  was found by the determined concentrations of  $^{41}\text{K}$  (also by epithermal neutrons) in the bulk material. Each gram of total potassium yields 30 Bq of  $^{40}\text{K}$ .

We have established a very reliable methodology to determine these radionuclides in typical edible food, geological specimens and tobacco. Table one shows the different irradiation, decay and counting times while Figure 1 shows the 74-keV gamma ray belonging to  $^{239}\text{U}$

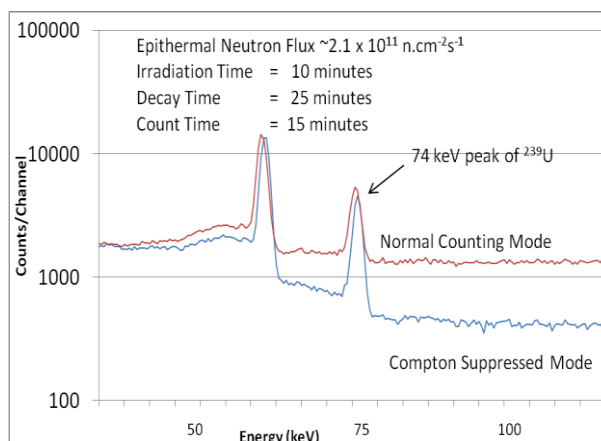


Figure 1 Typical gamma-ray spectrum of environmental, geological or food sample irradiated with epithermal neutrons showing the 74-keV gamma ray.

**Table 1 Irradiation Conditions for Neutron Activation Analysis**

Element	Neutron Energy	Irradiation Time	Decay Time	Counting Time
uranium	epithermal	5 min	15 min	30 min
potassium	epithermal	5 min	15 min	30 min
thorium	thermal	2 h	2 w	10 h

# <sup>137</sup>Cs Retainment in Central Greece Soils: A Study Undertaken a Radiocesium Halflife After Chernobyl

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Keywords: Cs-137, soil cores, Chernobyl, Central Greece

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The Chernobyl nuclear accident in 1986 has affected a large part of Greece, especially the Northern and Central Greece areas (Alexandropoulos 1986). Highest concentrations of radiocesium in soils were recorded in Grevena/Kalambaka areas ( $A_{\max} = 149 \text{ kBq/m}^2$ ) during an extensive and systematic study that covered the majority of Greece's soils (Simopoulos 1989). The highly afflicted areas (Fig. 1) were examined again at the 10-yr mark, and <sup>137</sup>Cs was found less ( $A_{\text{avg}} = 62 \text{ kBq/m}^2$ ), but still significant, compared to the initial values (Arapis 1999). The estimation of <sup>137</sup>Cs levels have been the main object of several studies in the Greek environment, however, the areas with the highest concentration have not been studied again since 1996.

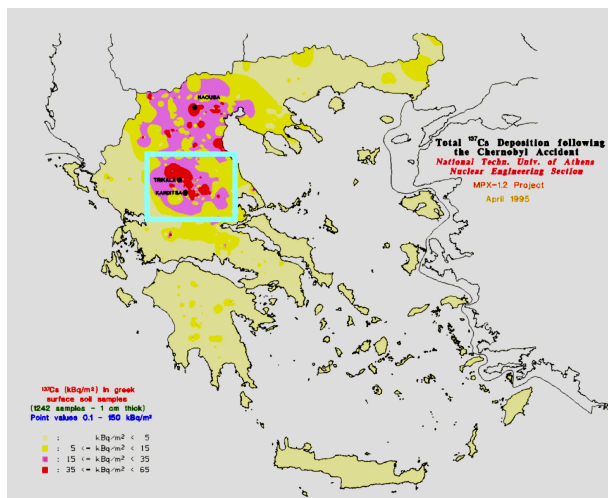


Figure 1. Total <sup>137</sup>Cs deposition following the Chernobyl Accident (Simopoulos 1989). The square inset marks the highly affected zone and target area of the present study

As the time from the fallout approaches one halflife of radiocesium ( $t_{1/2} = 30.08(9) \text{ y}$ ), new data are required to assess the long-term fallout impact on the Greek environment and its retainment in soils.

For that purpose, soil cores to a depth of 20 cm from ground surface were collected from several rural locations in Central Greece at the highly afflicted regions by the fallout. After thorough preparation (drying, sieving etc) the samples were examined by high-resolution gamma spectroscopy.

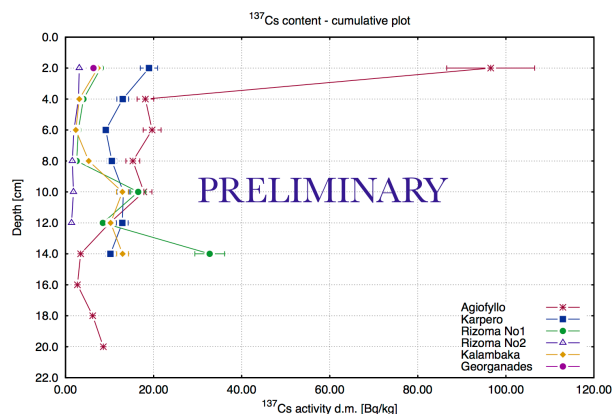


Figure 2. Preliminary results of <sup>137</sup>Cs depth distribution in soil cores (present study)

The results show that <sup>137</sup>Cs from Chernobyl is still highly present at some remote, undisturbed areas, while it has reached background levels or moved deeper into the soil profile in other locations (Fig. 2). Correlations of <sup>137</sup>Cs to the soil particle-size fraction have also been searched, especially focusing on the clay content. In addition, natural radioactivity levels have been examined.

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# Assessment of the external exposure of the Brazilian population to natural background radiation

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**Keywords:** external exposure, Brazil, natural background radiation.

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After the development of the GEORAD project (Silva et al, 2013), aiming to collect data on environmental concentrations in Brazil, a first trial of mapping Brazilian exposure to natural background radiation was attempted. Most data gathered up to now, however, reflect high background areas. Current surveys by car started to change this paradigm and gives an indication on exposure of overall population. Most areas surveyed by car up to now are related to coastal sites, where most of Brazilian population lives (Magalhães et al, 2015).

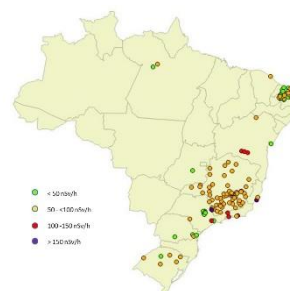
Exposure to cosmic radiation was calculated by the model proposed by UNSCEAR (2008), for ionizing and neutron components, based on latitude and altitude data. In a first approach, all municipalities with more than 100,000 inhabitants were included, comprising 253 counties and about 52% of the total Brazilian population. However, the area coverage was small, reflecting the uneven population distribution in the country (Figure 1).. Average contribution to the Brazilian population was estimated as 0.37 nSv/h, very close to the worldwide average estimated by UNSCEAR (2008) of 0.38 nSv/h.



Figure 1. Results for outdoors exposure to cosmic radiation (nSv/h)

For terrestrial radiation, two types of data were used: (i) open field gamma dose rate measurements; and, (ii) measurements of natural radionuclides in soil samples. Only results containing results for k-40 and at least one long lived nuclide of both U-238 and Th-232 series were included. Secular equilibrium in soil was considered and terrestrial contribution to external dose rates were calculated using the model proposed by UNSCEAR (2008). Only about 21 % of the population and 16 % of the total area of Brazil is covered with measurements that allows estimates of terrestrial external exposure. Average dose rate was estimated to

be 41.3. nSv/h. Results based on soil concentration were higher than those from gamma dose rate surveys. This was expected as most works describing soil concentration were done in high background areas while some of the more recent surveys are mostly related to densely populated areas. However, with the continuity of these long range car surveys, it is expected that the population related average external dose rate shall be lower than current values due to the small contribution of high background areas to overall Brazilian population.



# Observation and Modelling of Atmospheric Radon-222 Transport in East Asia

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*Keywords:* radon-222, atmospheric transport model, surface air concentration, WRF, East Asia

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## Introduction

Atmospheric  $^{222}\text{Rn}$  concentration, as a tracer for atmospheric transport, has been measured at several sites in our observation network in the East Asian region. In addition, a long-range atmospheric  $^{222}\text{Rn}$  transport model has been developed. It was pointed out in the model verification study using the surface  $^{222}\text{Rn}$  concentration measured by the network that, although the performance of this model was generally satisfactory, the model had a tendency of underestimating the surface concentration especially in winter. The purpose of this work is, therefore, to investigate the reasons of the underestimation by discussing transport processes based on observed data and model simulation results.

## Measurement of $^{222}\text{Rn}$ concentration in air

Atmospheric  $^{222}\text{Rn}$  concentration was measured at Hachijo (33.15°N, 139.75°E), which is a small island in the Pacific Ocean located 200 km to the south of the main island of Japan. The measurement was made with an electrostatic radon monitor, which has a detection limit of 0.5 Bq m<sup>-3</sup> for an hourly concentration and the accuracy is about 11% at 5 Bq m<sup>-3</sup>. Because of the small area and the geology of the island, locally exhaled  $^{222}\text{Rn}$  has already been evaluated to be negligible.

Figure 1 shows monthly average of  $^{222}\text{Rn}$  concentration at Hachijo.  $^{222}\text{Rn}$  concentration has a tendency to be high in winter and low in summer. The high concentration in winter has been found to be caused by continental  $^{222}\text{Rn}$  transported to Hachijo by the northwestern winter monsoon. Therefore, to investigate long-range atmospheric transport process of  $^{222}\text{Rn}$ , temporal variations in the concentration observed in winter were analyzed in this study.

## Long-range $^{222}\text{Rn}$ transport model

The three-dimensional long-range atmospheric transport model consists of two models, the Weather Research and Forecast (WRF) Model and an Eulerian atmospheric transport model. The calculation was conducted for the period from January 1 to 31, 2008, using a 9792 km × 7776 km domain covering East Asia. The calculation result of surface  $^{222}\text{Rn}$  concentration is compared with observation result in Figure 2. The monthly averages of the measurement and model calculation are 2.3 Bq m<sup>-3</sup> and 1.7 Bq m<sup>-3</sup>, respectively, indicating the underestimation in winter.

## Causes of underestimation

Causes of the underestimation were discussed separately for the three periods shown in Figure 2. As for the Period 1, the analyses of upper air meteorology revealed that the higher measured concentration than the calculation was maintained by an inflow of high concentration air at the northwest corner of the domain, followed by downward transport over the Pacific Ocean. The test calculation shown in Figure 2 with a hypothetical inflow of  $^{222}\text{Rn}$  at the boundary showed an

improved model performance. This result indicates that it is highly likely that the  $^{222}\text{Rn}$  concentration in upper air was as high as 1 Bq m<sup>-3</sup> or higher.

During Period2, there was a contribution of  $^{222}\text{Rn}$  from southern China. It was found by the analysis of vertical temperature profile that, along the transport path from southern China to Hachijo, the model slightly poorly calculated the atmospheric stability, causing excessive dilution of the surface  $^{222}\text{Rn}$  concentration due to vertical mixing.

Period 3 is characterized by a passage of a cold front near Hachijo. It was inferred that a steep horizontal concentration gradient across the front was formed and the model failed to reproduce the location of front and the microstructure of wind velocity field around the front.

## Conclusion

It was shown that  $^{222}\text{Rn}$  at higher altitude over the Asian continent significantly contribute the relatively high concentration over the Pacific Ocean in winter via long-range atmospheric  $^{222}\text{Rn}$  transport. Multiple causes were identified for the underestimation of surface concentration in winter.

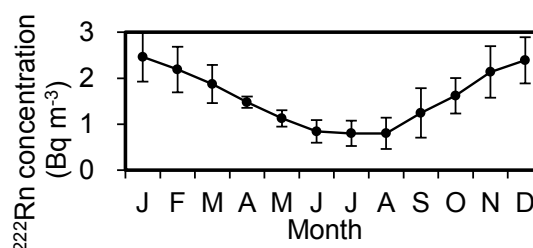


Figure 1, Monthly average of radon concentration at Hachijo-island during the 12 year period from 2001. Error bars represent standard deviation of year-to-year variation in monthly averages.

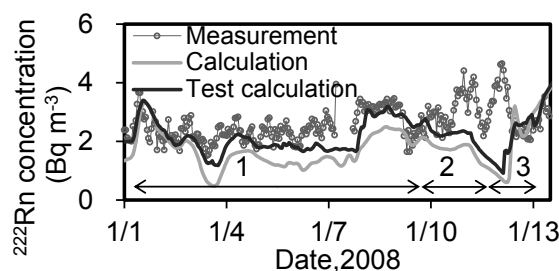


Figure 2, Comparison of temporal variation of surface  $^{222}\text{Rn}$  concentration in January 2008 between measurement and model calculation at Hachijo.

## Acknowledgments

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# Radiation hazard indices in the application of phosphogypsum mixtures as a building material: proposal for a regulation

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*Keywords: radiation hazard index, phosphogypsum, building materials, NORM.*  
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Phosphogypsum (PG), a waste by-product derived from the production of phosphoric acid, is being worldwide stockpiled, posing concerns about the environmental problems originating from this practice. Considerations about the viability of the safe reuse of this material have been raised, among them its potential use in civil construction. However, as PG can contain natural radionuclides in significant concentrations, using it as a building material has radiological implications, which have prevented such application.

Recently, a working group was established at the national regulatory level in Brazil, aiming to define a policy for using PG in this way. The adopted approach was to limit the concentration of PG to be mixed with natural gypsum, based on  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations found in PG.

In this study, the procedure described by Steger et al. (1992) to assess the hazard indices is employed, taking as input data concentration of Ra-226, Th-232 and K-40 in the material, radon exhalation fraction ( $\epsilon$ ) from the internal walls, the density ( $\rho$ ) and thickness ( $d$ ) of the walls.

The following values were adopted for the assessment:  $\epsilon = 0.1$ , which is a realistic figure considering that only 10% of the radon is emanated from the walls (Steger et al., 1992; Bossew, 2003);  $d = 0.1\text{m}$ , since this figure is above of the maximum admissible thickness for gypsum plates, according to Brazilian regulation, which is 15 cm. This value is equal to the width of the only gypsum brick model currently found to be produced in Brazil, compliant to the applicable engineering regulation.

For the density,  $\rho = 1750\text{ kg.m}^{-3}$ , that is the greatest density experimentally found for gypsum bricks, was assumed, and is also above of the maximum density allowed for plasterboard, which is  $933\text{ kg.m}^{-3}$ .

In the present study, radionuclides giving rise to gamma rays, by itself or by its progeny, were selected. In order to apply the assessment procedure, several hypothetical mixtures of PG and mineral gypsum were assumed, varying the PG percentages  $p$  in the mixtures.

The hazard indices were assessed for the following  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations in PG, as follows:

$^{226}\text{Ra}$ : 50, 100, 150, 200, 300, 400, 600, 800, and  $1000\text{ Bq.kg}^{-1}$ .

$^{228}\text{Ra}$ : 50, 100, 200, 300, and  $400\text{ Bq.kg}^{-1}$ .

For each  $p$  value, the activity concentrations of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the mixture were calculated.

In adopting such values, the criterion of fixing the upper limits of each range in values above of the typical experimental values from each main PG stockpile in Brazil was employed, in order to allow a well defined classification of PG in each range.

For mineral gypsum, the following fixed values were adopted, based in previous experimental data:  $2.8\text{ Bq.kg}^{-1}$  for  $^{226}\text{Ra}$ , and  $1.7\text{ Bq.kg}^{-1}$  for  $^{228}\text{Ra}$ . For  $^{40}\text{K}$  concentration,  $50\text{ Bq.kg}^{-1}$  was adopted for all PG/gypsum mixtures, as no analyzed PG in Brazil presented concentration above this value (Nisti et al., 2013).

Allowable PG percentage adopted, for each  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations pair, was the highest possible so that the resulting effective dose was below  $1\text{ mSv}$ .

Based on these assumptions, it was concluded that PG from the largest Brazilian deposits, could be used, from the radiation safety point of view, in all the range of percentages from 0% to 100%. In the extreme situations, being  $C(^{226}\text{Ra})$  and  $C(^{228}\text{Ra})$  the activity concentrations of the respective nuclides, PG should not be mixed for use at all if  $C(^{226}\text{Ra}) > 1000\text{ Bq.kg}^{-1}$ , and could be used in a percentage as high as 100%, in the following conditions:

$C(^{226}\text{Ra}) \leq 100\text{ Bq.kg}^{-1}$  and  $C(^{228}\text{Ra}) \leq 50\text{ Bq.kg}^{-1}$ ; or  
 $C(^{226}\text{Ra}) \leq 50\text{ Bq.kg}^{-1}$  and  $C(^{228}\text{Ra}) \leq 100\text{ Bq.kg}^{-1}$ .

These results show that the use of phosphogypsum in civil construction, especially mixed with natural gypsum, is actually viable and should be carefully considered and taken into account, in view of the positive impact on the environment by reducing the PG deposits.

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# The character of tritium contamination of the soil in the places of realization of the nuclear tests

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Keywords: «Experimental field» site, «Balapan» site, tritium, europium, method of autoclaved decomposition  
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During research of the territory of Semipalatinsk Test Site (STS) tritium was found in different environmental objects – surface and ground waters, vegetation, air environment, snow cover. Analysis of obtained data have shown that, contamination of environmental objects with tritium at the STS territory is associated with places of underground nuclear tests. It was supposed that during surface nuclear tests, mainly all the tritium was emitted into the atmosphere and then transferred with air streams beyond the test site. The decision was made to investigate the soil in places of surface and excavation explosion.

Aim of the work is to assess contamination of soils at the places of excavation nuclear tests with tritium.

As objects of research at «Experimental Field» site supposed explosion epicenters were chosen. At «Balapan» test site the crater of excavation explosion and external water reservoir, formed as a result of tests were taken as objects of research.

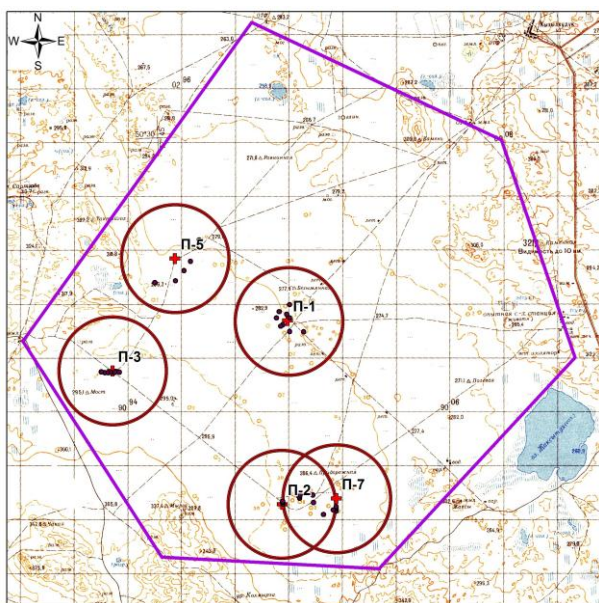


Figure 1. Location of sampling points of the soil at the site "Experimental field".

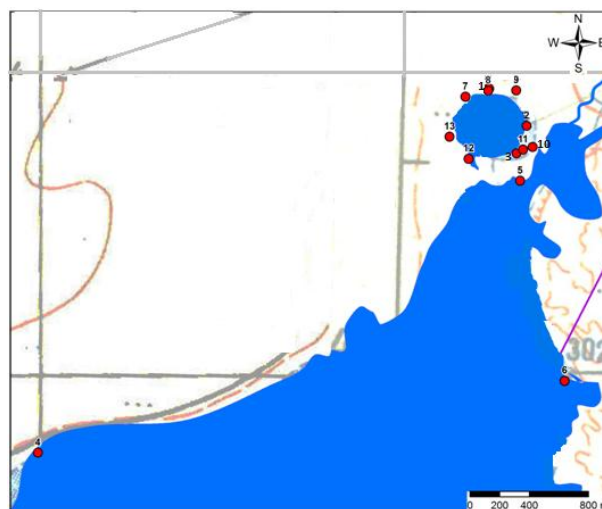


Figure 2. Location of sampling points of the soil at the site "Balapan".

As a result of research performed at «Experimental Field» site maximal values of specific activity of tritium in soil were found at the P-1 and P-5. Sites P-2 and P-7 are distinguished by lower level of soil contamination with tritium. The smallest concentration of tritium in soil was found at the P-3 site.

At «Balapan» site concentration of tritium contained in soil in significant amount. At the bank of external water reservoir detectable amount of tritium was found only in one point, which is the closest to the explosion crater. Concentration of tritium in soil at the rest two research sites was below the detection limit.

During performance of works it was found that concentration of  $^3\text{H}$  in soil correlates with concentration of  $^{152}\text{Eu}$ . Probably, concentration of tritium in soil depends on character and yield of charges used in the tests conducted.

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# Assessment of annual effective dose from $^{40}\text{K}$ by ingestion of food in Korea

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**Keywords:** potassium-40, annual effective dose, daily potassium intake

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It is well-known that  $^{40}\text{K}$  contributes to a large portion of annual effective dose by the ingestion. Many studies on the estimation of the internal dose received from  $^{40}\text{K}$  by measuring its radioactivity in foodstuffs have been conducted. But the kind of food sample has been restricted. In the purpose of obtaining comprehensive data of annual internal dose by consumption of  $^{40}\text{K}$  in many kinds of food, the  $^{40}\text{K}$  radioactivity in a daily intake of food was calculated by using statistical data for the amount of daily potassium intake and activity of  $^{40}\text{K}$  of elementary K. And then the annual effective dose from  $^{40}\text{K}$  by ingestion was estimated.

The equations for calculating the annual effective doses ( $\mu\text{Sv a}^{-1}$ ) by ingestion of  $^{40}\text{K}$  in foodstuffs or food groups are followed;

$$E = DCF \times I \times A \times 365$$

$$A = \frac{Na \times a \times \ln 2}{100 \times MW \times T_{1/2}}$$

where DCF is dose conversion factor,  $0.0062 \mu\text{Sv Bq}^{-1}$  provided in ICRP 74.  $I$  ( $\text{g day}^{-1}$ ) is the amount of potassium intake a day from annual report of Ministry of Health and Welfare of Korea.  $A$  ( $\text{Bq g}^{-1}$ ) is activity of  $^{40}\text{K}$  of elementary K.  $Na$  is Avogadro's number,  $6.022 \times 10^{23}$ .  $a$  is natural abundance of  $^{40}\text{K}$ , 0.0117%.  $T_{1/2}$  is half life of  $^{40}\text{K}$ ,  $1.28 \times 10^9$  year.  $MW$  is molecular weight of K,  $39.1 \text{ g mol}^{-1}$ . And total annual internal dose ( $\mu\text{Sv a}^{-1}$ ) by ingestion of  $^{40}\text{K}$  was obtained by summing up those from each food group. The results are listed in table 1, 2.

Table 1. The amount of daily intake of potassium in foodstuffs mainly contributing to effective dose among the most-consumed foodstuffs and the annual effective doses from those

foodstuff	amount of intake [mg day <sup>-1</sup> ]	effective dose [ $\mu\text{Sv a}^{-1}$ ]
polished rice	272.3 ± 3.6	19.07 ± 0.25
cabbage kimchi	127.7 ± 4.0	8.94 ± 0.28
milk	113.4 ± 3.4	7.94 ± 0.24
pork	103.7 ± 4.2	7.26 ± 0.29
potato	100.5 ± 7.5	7.04 ± 0.53
oriental melon	95.2 ± 12.0	6.67 ± 0.84

As shown in the tables, grain (polished rice), vegetables (cabbage kimchi) have a significant contribution to annual internal dose. It might be related to Korean dietary culture. The total annual internal dose

by  $^{40}\text{K}$  is approximately  $0.207 \pm 0.002 \text{ mSv}$  which is higher than worldwide value, 0.17 mSv, however it is within the typical range of the internal dose by ingestion from 0.2 mSv to 0.8 mSv.

Table 2. The amount of daily intake of potassium in food groups and the annual effective doses from those

food group	amount of intake [mg day <sup>-1</sup> ]	effective dose [ $\mu\text{Sv a}^{-1}$ ]
grain	487.6 ± 4.6	34.14 ± 0.32
potato, starch	187.8 ± 9.4	13.15 ± 0.66
sugars	15.0 ± 1.7	1.05 ± 0.12
pulses	90.6 ± 3.2	6.34 ± 0.22
seeds	25.4 ± 1.4	1.78 ± 0.10
vegetables	722.4 ± 11.1	50.58 ± 0.78
mushroom	14.8 ± 0.9	1.04 ± 0.06
fruits	347.7 ± 12.7	24.35 ± 0.89
seaweed	109.7 ± 4.1	7.68 ± 0.29
drinks	132.2 ± 3.8	9.26 ± 0.27
liquors	19.2 ± 1.1	1.34 ± 0.08
spicery	179.3 ± 3.2	12.56 ± 0.22
fat and oils(vegetable)	20.9 ± 1.1	1.46 ± 0.08
others(vegetable)	12.0 ± 2.2	0.84 ± 0.15
meat	271.8 ± 8.7	19.03 ± 0.61
eggs	40.9 ± 1.1	2.86 ± 0.08
seafood	119.0 ± 3.7	8.33 ± 0.26
milk	164.3 ± 4.5	11.50 ± 0.32
fat and oils(animal)	0.1 ± 0.0	0.01 ± 0.00
Others(animal)	0.2 ± 0.1	0.01 ± 0.01
Total	2961.0 ± 30.3	207.34 ± 2.12

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# Origin of halides (Cl<sup>-</sup> and Br<sup>-</sup>) and of their stable isotopes ( $\delta^{37}\text{Cl}$ and $\delta^{81}\text{Br}$ ) at the Tournemire clayrock URL (France) - Experimental and numerical approach

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**Keywords:** Radioactive waste, Clay rock, natural tracers, cubic diffusion, porewater.

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This work is part of research conducted by the Institute of Radiation Protection and Nuclear Safety (IRSN) on the geological disposal of High-Level and Intermediate-Level Long-Lived (HL-ILL) radioactive waste in deep clayrocks. In France, the choice of the potential host rock for the geological storage is focused on the Callovian-Oxfordian (COx) of Meuse/Haute-Marne because of its low permeability, capacity for self-sealing, high sorption and capacity of transport of radionuclide (RN) by diffusion, whose studies are driven mainly by the ANDRA (Agence Nationale pour la gestion des Déchets RADIOactifs). IRSN, which plays an expert role for ASN (l'Autorité de Sûreté Nucléaire), has its own underground research laboratory in a clayrock which has strong analogies to the COx. This is the Toarcian/Domerian clayrock located at Tournemire in southern Aveyron in France.

The purpose of this work was to assess the transfer of RN in the Tournemire clayrock through the study of halides contents and of their stable isotopes (Cl<sup>-</sup>, Br<sup>-</sup>,  $\delta^{37}\text{Cl}$ ,  $\delta^{81}\text{Br}$ ). The multiple approaches used consisted for halides to: 1) Assess their stock in different fractions of the rock by applying several techniques including i) alkaline fusion for their total stock, ii) leaching to access their stock in porewater and to mineral phases sensitive to dissolution iii) cubic diffusion for their stock in porewater, 2) Get their diffusive transport parameters of a selection of samples from the upper Toarcian by cubic diffusion experiments modeled using the Hytec transport code developed by Mines ParisTech, (van der Lee, 2007) and 3) Model their transport after palaeohydrogeological known changes of the Tournemire massif, (Peyaud, et al., 2005).

The experimental approach, conducted at the LAME lab (Laboratoire d'Analyses et des Moyens Expérimentaux, France), did not lead to an operational protocol for the alkaline fusion due to an incomplete rock dissolution. Leaching was used to characterize the concentrations of halides in the fractions of pore water and of minerals sensitive to dissolution. The results show concentrations of halides much higher than those of pore water with very low Cl/Br ratios likely resulting from the dissolution of mineral species. The cubic diffusion produced the pore diffusion coefficients for Cl and Br as well as their concentration in the pore water. Cubic diffusion also allowed estimating a Cl to Br pore diffusion coefficient ratio, necessary to calculate the profiles of Cl/Br. These estimates have required the use of the transport code Hytec i) for dimensioning and implementing the experiment in a time frame compatible

with the work period, ii) for analysing the sensitivity of the model to the accessible porosity and to the diffusion coefficient which act respectively to the steady phase and transient phase of the experiments, and finally, iii) for adjusting the pore diffusion coefficients of Cl and Br to an accessible porosity of 3-4% (Patriarche, D., 2004).

The Hytec code was then used to check the consistency of the current profiles of chlorides, bromides,  $^{35}\text{Cl}$ ,  $^{37}\text{Cl}$ ,  $\delta^{37}\text{Cl}$ , Cl/Br in 1D, a fake drilling assumed crossing the entire clayrock. The assumption is that halides have undergone a diffusive transport between seawater trapped during sedimentation and meteoric waters infiltrated at different times to domain boundaries. Four scenarios were tested according to the paleohydrogeological history of the massif. All tracers and scenarios are consistent with a unique marine source of halides more or less diluted by meteoric waters. The duration of the diffusive exchange initially suggested 85 ± 10 Ma (Bensenouci, 2010) is never contradicted despite uncertainties related to changes in boundary conditions. This body of evidence would suggest that molecular diffusion is the transport process which has affected and still affect the Tournemire clayrock, outside of fault zones. The  $\delta^{37}\text{Cl}$  results expected on the surrounding carbonated aquifers, leachates and fracture waters (including  $\delta^{81}\text{Br}$  values; data processing in progress) should help to refine the models and the results.

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# Effect of phosphogypsum amendments in the U and Th uptake and partitioning in cotton crops grown in reclaimed soils from the Guadalquivir marshlands (SW Spain)

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**Introduction:** In the estuarine region of the Guadalquivir River, agriculture soils are saline and sodic. They have been treated for decades with Ca amendments, mainly phosphogypsum (PG) from the phosphate industries in the nearby city of Huelva. PG is composed mainly  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ , but with some levels of fluoride and certain natural-occurring radionuclides, such as U and  $^{226}\text{Ra}$ . Thus, the agriculture use of PG can introduce bio-available U in soils, from where it could be transferred into the food chain. This work reports results from a field experiment conducted in a commercial farm in the marshland area of Lebrija, SW Spain.

**Method:** The experimental site ( $37^\circ 1.2' \text{N}$ ,  $6^\circ 7.4' \text{W}$ ) was divided into six plots of 0.5 ha ( $20 \text{ m} \times 250 \text{ m}$ ), with tile drains placed at 1 m below the soil horizon, longitudinally distributed and spaced every 5 m. Two treatments were designed, control C, and PG ( $25 \text{ t ha}^{-1}$  PG, Table 1), both in triplicate. Abril *et al* (2008).

Before seeding cotton,  $2000 \text{ kg ha}^{-1}$  of NPK fertilizer (12% N, 10% P, 30% K) was applied as basal fertilizer, Table 1). During the growing process, cotton was irrigated seven times with drainage, Table 1. Soil and plant samples were prepared by procedures described by Enamorado *et al* (2013), and then measured by ICP-MS.

Table 1. Th and U concentrations in PG, NPK fertilizer, irrigation water and non-reclaimed soil.

Inputs ( $\text{mg kg}^{-1}$ )	Th	U
PG	$2.48 \pm 0.04$	$16.2 \pm 0.5$
NPK	$1.21 \pm 0.07$	$49 \pm 3$
Irrigation water	$0.028 \pm 0.07$	$1.35 \pm 0.08$
Non-reclaimed soil	$6.8 \pm 0.3$	$1.64 \pm 0.06$

\*The irrigation water is measure in  $\mu\text{g L}^{-1}$ .

**Results:** Considering the Th in farm soil ( $7.8 \pm 0.3$ )  $\text{mg kg}^{-1}$ , the inputs were lower than 1% of the amount in the top 0-40 cm soil layer. For U, the amount introduced through the NPK fertilizer was 1% of the total inventory, while PG increased it by a ~6%. Finally, additional U was introduced through the irrigation waters, with a net gain of ~6%, when taking into account losses through drainage.

Although, differences between C and PG treatments were not statistically significant in farm soils during the experiment. After 30 years of PG applications, U concentrations in farm soils (Table 2) were significantly higher than those found in the nearby non-reclaimed soils. Table 2 shows that U-concentration in plants from PG-amended plots were statistically higher than those found in C-plots. Finally, U-losses by drainage were significantly higher in PG plots (Table 2).

Figure 1 shows how U-concentrations in drainage water follow a logarithmic trend of decrease with instantaneous water flow for both treatments.

Table 2. U concentrations in farm soils after PG application, in harvested cotton plants, and total U losses by drainage.

	C	PG
Soil ( $\text{mg kg}^{-1}$ )	$2.88 \pm 0.07$	$3.21 \pm 0.19$
Cotton plant ( $\text{mg kg}^{-1}$ )	$0.058 \pm 0.016$	$0.16 \pm 0.12^*$
U in drainage (kg)	$1.03 \pm 0.02$	$1.36 \pm 0.04^*$

Mean and standard deviation of mean. Values followed by \* are significantly higher (Tukey's test at 95% CL).

**Conclusions:** The application of PG as a soil Ca-amendment at the usual rate in the area, of  $25 \text{ t ha}^{-1}$ , and with the typical agricultural practices, significantly increased the U uptake by cotton plants and the total U-losses through the drainage waters. Although U-concentrations in the 0-30 cm soil horizon did not show statistically significant differences between both treatments, the cumulative effect of PG application over three decades is well revealed when compared with nearby non-reclaimed marsh soils. As the uptake may depend on the crop, it would be desirable to extend this study to other commercial crops grown in the area to prevent any potential health hazards.

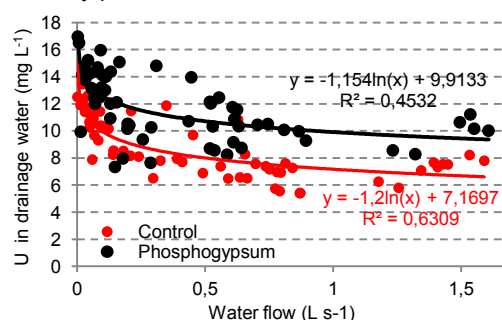


Figure 1. U concentrations in drainage waters.

## Acknowledgments

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# Groundwater recharge of injection-pumping system for water-curtain heating system using radon mass balance

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Keywords: water-curtain cultivation, green-house, groundwater recharge, radon mass balance  
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Groundwater has been widely used as a thermal source in water-curtain heating systems at the green-house cultivation area during winter season in Korea. So far, groundwater is extracted through the pumping well and then used in the water-curtain heating system; finally the used groundwater is drained into a stream. Because groundwater has been over-extracted for the water-curtain heating system, most green-house cultivation area has been suffering from groundwater shortage problem.

For the restoration of the groundwater level, a large-caliber injection-pumping system (IPS) was examined in this study (Fig. 1).

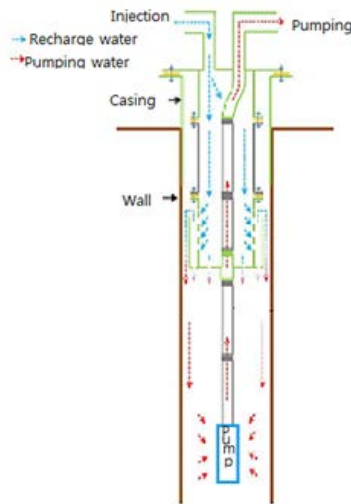


Fig. 1. Schematic of the IPS.

The IPS consists of an injection pipe and a pumping pipe with a submersible water pump in a large outer pipe. The used groundwater was collected into a collecting tank, and then sent into the recharge-pumping system through the injection pipe.

Radon (Rn-222) as a natural tracer was applied to evaluate recharge efficiency of the IPS. Radon activity concentrations of pumping groundwater and used groundwater were about 28,000 Bq/m<sup>3</sup> and 5,000 Bq/m<sup>3</sup>, respectively. A continuous radon monitoring system was used to measure radon activity continuously in pumping water from the IPS with and without injection of used groundwater (Fig. 2).

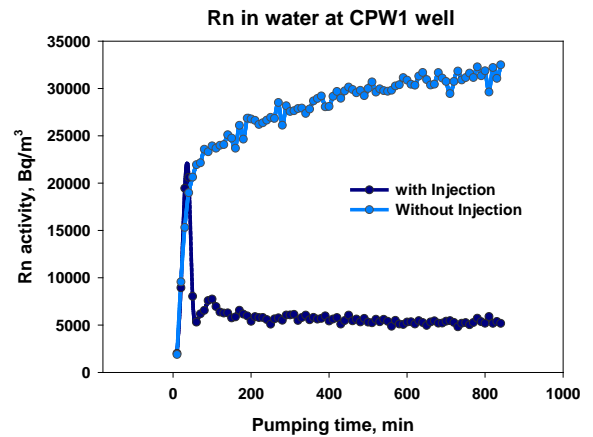


Fig. 2. Radon activity concentrations in pumping water of RPS with and without injection used groundwater.

A radon mass balance equation was derived from radon activity of intrinsic groundwater (g), injected used groundwater (i) and mixed groundwater (m), as (1).

$$A_m V_m = A_g V_g + A_i V_i, \quad V_m = V_g + V_i \quad (1)$$

A and V represent radon activity concentration (Bq/m<sup>3</sup>) and water volume (m<sup>3</sup>), respectively. Recharge efficiency can be denoted by the mixing ratio,  $V_i/V_g$ , (2).

$$\frac{V_i}{V_g} = \frac{A_m - A_g}{A_i - A_m} \quad (2)$$

As shown in radon mass balance equations (1) and (2), the recharge efficiency of our IPS could be estimated using radon activity concentrations of fresh groundwater, injected used groundwater and mixed groundwater.

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## BIOLOGICAL SITES FOR MONITORING OF RADIATION HAZARDOUS OBJECTS

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**Keywords:** radioecology,  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ,  $^3\text{H}$ , Semipalatinsk Test Site.

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Results of several years of research in the area of redistribution of artificial radionuclides in the «soil-plant» system as well as study of possibility of use of plants as indicators of  $^3\text{H}$  radionuclide concentration in ground water in the territory of Semipalatinsk Test Site (STS) (Larionova, 2010, 2013) summarize to conclusion of creation of biological monitoring sites for radiation-hazardous objects. The most widely used methods of radioecological monitoring (systems of hydrogeological boreholes and air monitoring posts) reflect the situation to exact period of time but they are not always able to assess integrally the over-past period. From this point of view biological monitoring sites (represented by certain groups of plants of definite species composition) are able to characterize radiation situation for definite period of time. That is related to life duration of both individual species of plants and their individual organs. Another advantage in use of biological monitoring sites is simplicity of realization and economic benefit (they don't need continuous service and energy resources).

Research was conducted on the STS territory. There still exists principal possibility of radionuclides transfer with water from the underground places, where nuclear tests have been performed on the «Degelen» site. The objects for research places have been selected beyond the «Degelen» site where the land surface and ground water exit.

In order to find ratios and mechanisms of redistribution of radionuclides in different environmental components the following field works have been performed: drilling of hydrogeological boreholes; laying of soil sections and environmental sampling (water (land surface and ground water), soil, vegetation). The following plant species were analyzed: hydrophytes (*Phragmites australis*, *Carex melanostachya*, *Calamagrostis arundinacea*), growing in the area of surface water stream, and phreatophytes (*Salix triandra*, *Populus nigra*), mainly using ground waters as the source of moisture.

Specific activity of radionuclides  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  was measured by gamma spectrometry; for  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$  method of radiochemical extraction with further beta and alpha spectrometry was used and for  $^3\text{H}$  – liquid scintillation spectrometry was used. Specific activity of  $^3\text{H}$  in plants was measured in their organic components and free water that is evaporated by plants.

The results of analysis specific activity of radionuclides in water are:  $^3\text{H}$  35000-75000 Bq/kg;  $^{90}\text{Sr}$  – <0.25-14 Bq/kg;  $^{239+240}\text{Pu}$  – <0.0002-0.002 Bq/kg. Values of quantitative contains of  $^{241}\text{Am}$  (<3 Bq/kg) and  $^{137}\text{Cs}$  (<0.03 Bq/kg) in water was not determined.

Maximal specific activity of radionuclides ( $^{241}\text{Am}$  – up to 7 Bq/kg,  $^{137}\text{Cs}$  – up to 140 Bq/kg,  $^{90}\text{Sr}$  – up to 1200 Bq/kg and  $^{239+240}\text{Pu}$  – up to 35 Bq/kg) were noted in the top soil layer and decrease in the depth of soil profile. That can indicate radionuclides transfer with surface waters.

Specific activity of  $^3\text{H}$  in free water of plants is 10000-45000 Bq/kg, in organic components – 7900-17000 Bq/kg. Concentration of  $^{90}\text{Sr}$  in plants is 4.8-350 Bq/kg;  $^{239+240}\text{Pu}$  – <0.1-13 Bq/kg;  $^{241}\text{Am}$  – <0.4-2.6 Bq/kg;  $^{137}\text{Cs}$  – <1-6.7 Bq/kg. The fact that radionuclides values  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  of specific activity in plants is higher than in water could be connected with ability of plants to accumulate them and non-continuous transfer of radionuclides with water.

In this case plants can be indicators of radionuclides concentration in the soil cover and surface water. Moreover it could also characterize deeper soil layers, as well as ground water of respective territories and ground waters of respective territories. Specified values of specific activity for radionuclides  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  in plants indicate possibility to use them for assessment of waters and soils during monitoring of radiation-dangerous objects.

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## Preliminary results of $^{238}\text{U}$ , $^{232}\text{Th}$ and trace elements determined in soil and sediment profiles collected in Ponte Nova Reservoir, São Paulo, Brazil

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Reservoirs worldwide have been built since the early XX century to 1970's and 1980's in order to supply water for populations and years later generate electricity. In Brazil, the majority of its reservoirs has the main objective of public supply of water, to control of the flow of rivers and generate electricity. The state of São Paulo, located in the Southeast region of the country has at least 15 reservoirs that are considered medium to large and are used to provide water for the biggest population of the country (Esteves, 2011).

The Tietê River, the most important river of the state of São Paulo, from its source in the city of Salesópolis to its mouth on the Parana River is dammed in several parts and form large artificial reservoirs. Ponte Nova reservoir, located in the cities of Salesópolis and Biritiba Mirim, is one the large reservoirs formed by the waters of Tietê River near of its source. It was the first reservoir built in this part of the river in 1972 in order to control the ebb of the Tietê River in the metropolitan region of São Paulo. Ponte Nova also, supplies water for the cities in its around, and especially for a large producing region of agricultural products for the city of São Paulo (Tundisi and Tundisi, 2008).

The watersheds that form an artificial reservoir, like Ponte Nova, interact with the soil around the reservoir and its sediment, through physical and chemical weathering. This interaction can change the quality of water from a reservoir, especially when these water bodies go through drought periods.

Hence it is important not only one monitoring the quality of water from a reservoir, but also to analyze its soil and sediment, because a reservoir is the final destination of all the alterations that may have occurred around.

Therefore the objectives of this work were to present preliminary results of the trace elements As, Ba, Br, Ca, Ce, Co, Cr, Cs, Eu, Fe, Hf, K, La, Lu, Na, Nd, Rb, Sb, Sc, Ta, Tb, Th, U, Yb and Zn, using Instrumental Neutron Activation Analysis (INAA). This technique has been extensively used in geochemical studies due to the possibility of multielemental analyzes at the same time with excellent precision and accuracy, without a previous process of sample digestion; the detection limits varied from 0.01 to 1 mg kg<sup>-1</sup> for most elements.

A soil profile and a sediment core were collected in the most important lithology of the reservoir in the upstream. The lithology of this reservoir has predominance of metamorphic rocks, calcsilicates and

amphibolites, with the presence of granite calcalkalines (Perrota et al, 2005).

The soil profile, of 1,2 m long, was cut each 5 cm and the sediment core, 50 cm long, each 3 cm. The samples, soil and sediment, were dried at room temperature, sieved in a 2.00 mm sieve, ground in a mortar, and then homogenized before analysis.

For INAA, about 150 mg of the sample and reference materials were accurately weighed and sealed in pre-cleaned polyethylene bags, and were irradiated for 8 hours, under a thermal neutron flux of  $1 \text{ to } 5 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$  at the IEA-R1 nuclear research reactor at IPEN. Two series of counting were made: the first, after one week decay where As, Br, Ca, K, La, Na, Nd, U, Sb, Sm and Yb were measured and the second, after 15-20 days where the others elements were measured. Gamma spectrometry was performed using a Germanium detector hyperpure (HPGe) with relative efficiency of 23% and resolution of 2.1 keV for the  $^{60}\text{Co}$ , INTERTECHNIQUE, with an associated electronics. Spectra were analyzed using the Winner Winner Inter Gamma the ORTEC 6.0 program. For methodology validation regarding precision and accuracy reference materials SL-1 (Lake Sediment – IAEA) and Montana II soils (NIST) were used (Damatto, 2010).

The results obtained showed a clear correlation between the radionuclides determined in the soil and sediment samples. The enrichment factor and geoaccumulation index were used as a geochemical tool to evaluate a probable contamination of the reservoir.

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# Screening of bacterial strains isolated from uranium mill tailings porewaters for bioremediation purposes

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Keywords: bacteria, uranium, bioremediation.  
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The present work consists in the isolation and characterization of bacterial strains from porewaters sampled in the vicinity of two French uranium tailing repositories. Since microbes are known to affect decisively uranium fate and behavior in these environments (Francis, 1998), their identification and characterization is considered the first logical step for their eventual biotechnological application (Gadd, 2000).

In total 32 different bacterial isolates were recovered and assigned to 13 bacterial genera (comprised in the phyla Firmicutes, Actinobacteria and Proteobacteria) through partial 16S rRNA gene sequencing. For the bacterial isolates screening, a multidisciplinary approach including enzyme profiling, carbohydrates utilization tests and heavy metal tolerance evaluation was performed. The bacterial strains Br3 and Br5, corresponding to *Arthrobacter* sp. and *Microbacterium oxydans*, were selected and demonstrated to possess the ability to immobilize uranium by using High Resolution Transmission Electron Microscope (HRTEM). Moreover, Scanning Transmission Electron Microscope-High-Angle Annular Dark-Field (STEM-HAADF) analysis showed U accumulates on the surface and within bacterial cytoplasm, in addition to the extracellular space (Figure 1). Energy Dispersive X-ray (EDX) element-distribution maps demonstrated the presence of U and P within these accumulates.

These results indicate the potential of certain bacterial strains isolated from porewaters of U mill tailings for immobilizing uranium. Some of these bacterial strains might be considered as promising candidates in the design of uranium bioremediation strategies.

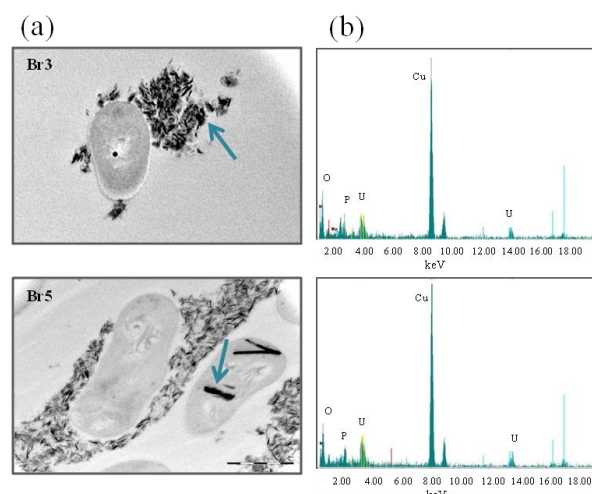


Figure 1. Transmission electron micrographs (a), coupled with energy dispersive X-ray spectra (b), of thin sections of the strains Br3 and Br5 treated with 0.5 mmol l<sup>-1</sup> of U(VI). The metal accumulated (indicated with blue arrows) is localized on the cell surface, intracellularly and also extracellularly as precipitates.

## Acknowledgments

This work results from a Joint Research Project between AREVA Mining Environmental R&D and the Department of Microbiology of the University of Granada. The authors thank AREVA Mining for financial support.

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# Hydrogeochemistry and natural radioactivity of a complex karst system: case study of Sierra de Gádor (Almería, SE Spain)

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Keywords: Uranium, <sup>226</sup>Ra, hidrogeochemistry, karst aquifer

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## Introduction

Due to the arid climate of the province of Almería, and the shortage and intermittency of its hydrographic network, the knowledge of the groundwater behavior is very relevant because it is the main source of supply of this resource for agricultural and urban use (Daniele et al., 2013)

The Sierra de Gádor is a complex karst aquifer system located in the southeastern Spain, and surrounds the town of Almería. It occupies an area close to 900 km<sup>2</sup> and supports some of the most important adjacent aquifers, the Bajo Andarax and the Campo de Dalías. The behavior of both major and trace elements has been little studied, specially the natural radionuclides.

The aim of this work was to evaluate the levels of trace elements and natural radionuclides (U isotopes and <sup>226</sup>Ra) in the Sierra de Gádor aquifer systems, trying to deepen in the knowledge of the basic processes that regulate the behavior of this system. In addition, we are going to carry out an evaluation of the radiological quality of the waters in the Sierra de Gádor.

## Materials and methods

During the month of June 2006, 35 representative points were selected; 28 groundwater points and 7 of surface water. At each point were collected 3 samples, two samples of 100 mL for the analysis of major, minors and trace elements, and another one of 6 L for the analysis of natural radionuclides (U isotopes and <sup>226</sup>Ra). The physicochemical parameters pH, redox potential (Eh), dissolved oxygen, temperature, electrical conductivity and HCO<sub>3</sub> contents were determined in situ.

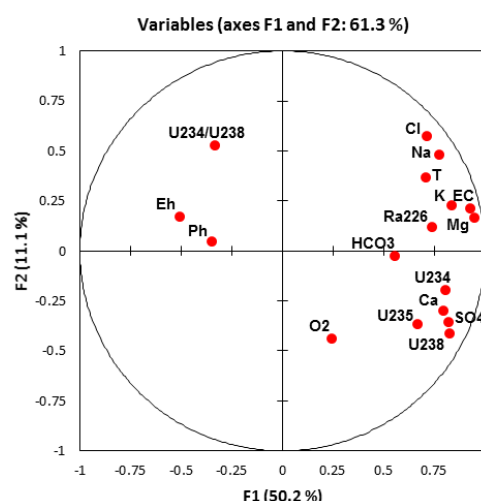
Analyses of major, minor and trace elements were carried out by combination of HR-ICP-MS and ICP-OES techniques. The activity concentration of the U isotopes and <sup>226</sup>Ra was determined using alpha-particle spectrometry with PIPS detectors.

## Results and discussion

In some points high contents of natural radioactivity are reached, especially in locations with high electrical conductivity and high Ca, SO<sub>4</sub> and Mg concentrations. The <sup>234</sup>U/<sup>238</sup>U activity ratios were found between 1.1 and 3.8 (with a medium value of 2.1), indicating this fact the existence of preferential leaching process along the aquifer and/or different contact times between water and aquifer materials (González et al., 2005). A Principal Component Analysis was applied in

order to correlate the radionuclides levels, physicochemical parameters and the major ions. (Figure 1).

Figure 1. Loading plot of variables.



The maximum effective dose from ingestion of the U isotopes and <sup>226</sup>Ra in these waters are far below the guideline value set to 100 μSv·y<sup>-1</sup> (Table 1), except for 3 locations. The main contributor to the effective dose for these waters is the <sup>226</sup>Ra, and by that is very recommendable the measure of this radioisotope in the radiological control plans of the water in this area.

Table 1. Concentrations and effective dose of the samples.

Isotope	Concentration (mBq/L)		Effective dose (μSv/y)	
	Max	Mean	Max	Mean
<sup>238</sup> U	44	25	1.4	0.6
<sup>235</sup> U	2.5	1.2	0.084	0.039
<sup>234</sup> U	80	40	2.9	1.4
<sup>226</sup> Ra	390	33	81	6.9

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# Impact of Radioactive Waste Discharges to the Irish Sea: A Study of Artificial Radioactivity Concentrations in Carlingford Lough, Ireland

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**Keywords:** radioactivity, marine, radiation pathway, waste discharges

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Low levels of radioactive effluent are routinely discharged into the north-east Irish Sea from nuclear reprocessing facility at Sellafield, UK. These discharges have given rise to enhanced levels of radioactivity in a wide range of environmental media such as sea water, marine sediments, seafood, seaweed and beach sediment particularly in the eastern Irish Sea.

A study of radioactivity in and around Carlingford Lough was carried out between 2011 - 2013. Carlingford Lough is a shallow estuary approximately 13 km in length and 5 km at its widest, with a relatively narrow stretch of ~1.7 km at the entrance. The shores of the Lough, lying along the border between Northern Ireland and the Republic of Ireland, range from rocky cliffs in the west to large expanses of mud flats in the east. At the mouth of the lough, the area is low-lying with sand and shingle beaches, with patches of mud and stones. Samples were collected at the locations shown on the map in Figure 1.

The study was conducted jointly by the Radiological Protection Institute of Ireland, now the Environmental Protection Agency (EPA), the Northern Ireland Environment Agency (NIEA) and University College Dublin (UCD).

This study repeats various aspects of the project originally performed in 1990 and reported in the first Carlingford Lough study (Mitchell et al, 1992). Changes in measured concentrations and doses are assessed.

The study included a dose assessment for high-rate seafood consumers living in the Carlingford Lough area. Seafood consumption data for the two critical groups (group A and group B) identified in an assessment of aquatic radiation pathways in Ireland (CEFAS, 2008) are used to perform the dose assessment.

A separate study of Carlingford Lough was conducted in 1997 which found increased concentrations of Tc-99 in the marine environment as a result of higher marine discharges of this radionuclide since 1994. This study assesses current levels of Tc-99 in the marine environment now that discharges have returned to pre-1994 levels. (Long et al., 1999).

Results of the current study are presented.

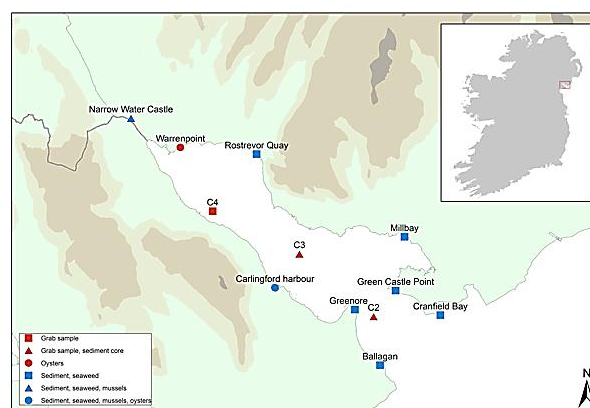


Figure 1. Location of sampling sites in Carlingford Lough, Ireland

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# A Study on the Levels of Radioactivity in Fish Samples from Experimental Lakes Area in Canada

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**Keywords:** natural radionuclides, caesium, freshwater fish, Fukushima impact.

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As part of Health Canada's study on background radiation levels in country foods, a total of 125 fish samples were collected from three lakes in the Experimental Lakes Area (ELA) during the summer of 2014: Lake 305 (11 Northern Pike, 20 Lake Whitefish); Lake 302 (20 White Sucker, 40 Lake Whitefish); and Lake 226 (34 Lake Whitefish). Naturally occurring radionuclides (such as Ra-226, Pb-210, Po-210 and K-40) as well as long lived contaminants (such as Cs-137) were analysed. Results are given in Bq/kg fresh weight (fw), as summarized in the Table below.

Results showed that Po-210 is the dominant contributor to radiation doses resulting from fish consumption. While concentrations of Pb-210 and Ra-226 were below conventional detection limits, Po-210 was measured in almost all fish samples collected from the ELA. The average concentration was about 1.5 Bq/kg fw. The resulting radiation dose from consumption of 1 kg of fish containing 1.5 Bq of Po-210 would be 2 µSv, a very small fraction (< 1/1000) of the annual dose (about 3 mSv) from exposure to natural background radiation in Canada.

Radioactive caesium-137 was measured using a new, more sensitive technique (Zhang et al. 2013). Cs-137 remains in the environment from the fallout of atmospheric nuclear weapons tests in the 1950s and 1960s as well as from nuclear reactor waste and accidental releases such as the Chernobyl accident in 1986 and the Fukushima accident in 2011. Cs-137 levels in contemporary samples were compared to data from samples collected from the same lakes in 1981 (Elliott et al. 1981). The mean Cs-137 concentration of ELA Lake Whitefish in 1981 was 52 Bq/kg fw (n=37). Lower levels in contemporary Lake Whitefish were predicted due to the physical half-life of Cs-137 (30 years). However, we observed an average Cs-137 concentration of 5.5 Bq/kg fw for all 94 samples of whitefish, much lower than that

predicted from physical half-life alone. Research projects are currently underway to study ecological half-lives and removal processes of Cs-137 in freshwater ecosystems.

Our analyses indicate that the average concentration of Cs-137 in fish from inland lakes is significantly higher than the observation in marine fish harvested from Canadian west coast in 2013 (Chen et al. 2014). However, it should be mentioned that the relatively high concentrations of Cs-137 in fish samples from inland lakes are still considered very low from radiological protection perspective. The resulting radiation dose for people from fish consumption would be a very small fraction of the annual dose from exposure to natural background radiation in Canada. Therefore, fishes from inland lakes, rivers and oceans pose no radiological health concern.

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LAKE	species	samples	Po-210 (Bq/kg fw)	Pb-210 (Bq/kg fw)	Ra-226 (Bq/kg fw)	Cs-137 (Bq/kg fw)	K-40 (Bq/kg fw)
305	Northern Pike	11	1.7 (±1.2)	< 0.2 (<DL, 0.2)	< 0.06	15.6 (±2.5)	62.8 (±6.1)
302	White Sucker	20	1.6 (±0.6)	0.4 (<DL, 0.7)	< 0.06	4.0 (±0.7)	75.9 (±8.7)
305	Whitefish	20	2.2 (±2.4)	< 0.2 (<DL, 0.4)	< 0.06 (<DL, 0.1)	4.9 (±0.4)	80.4 (±4.7)
226	Whitefish	34	2.1 (±1.3)	< 0.2	< 0.06 (<DL, 0.1)	5.5 (±0.6)	72.9 (±5.9)
302	Whitefish	40	0.5 (±0.3)	< 0.2	< 0.06	5.7 (±0.6)	87.1 (±4.3)



# Optimization of low-level liquid scintillation counter for $^{90}\text{Sr}$ determination in water samples using Cerenkov radiation

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Keywords:  $^{90}\text{Sr}$ , Cerenkov radiation, liquid scintillation counting

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$^{90}\text{Sr}$  is one of the important fission products likely to be found in the environment of a nuclear power plant or a fuel reprocessing plant. Due to its chemistry, similar to calcium's,  $^{90}\text{Sr}$  follows the paths of this element in the food chain and enters the human body. Radiostrontium is one of the most biologically hazardous radionuclides because it can enter the human body through the food chain, accumulates in bone tissue and its radiation causes damage to bone marrow. Because of its long physical and biological half-life the longest-lived radiostrontium isotope,  $^{90}\text{Sr}$  is of major concern in environmental contamination and nuclear processes.

If the energy of a beta particle, emitted by a radionuclide, is higher than 263 keV, this particle can produce Cerenkov radiation when it passes through water. The  $^{90}\text{Sr}$  beta spectrum has a maximum energy of 546 keV which is well above the Cherenkov threshold in pure water. More realistic is the Cherenkov counting of the  $^{90}\text{Y}$  beta spectrum which has a maximum energy of 2283 keV. This radionuclide is the daughter of  $^{90}\text{Sr}$  and has a half-life of 64.1 h and it is presumed that this pair is in secular equilibrium in environmental samples. Thus, by measuring  $^{90}\text{Y}$  activity in a sample, the  $^{90}\text{Sr}$  activity of the sample can be estimated.

Results of experiments were obtained using Ultra Low Level Liquid Scintillation Spectrometer Wallac Quantulus 1220<sup>TM</sup> manufactured by PerkinElmer (Finland, 2002), the spectra were acquired by EASYView and WinQ software. A standard radioactive source activity (aqueous  $^{90}\text{Sr}/^{90}\text{Y}$  solution) produced from Czech Metrology Institute, Inspectorate for Ionizing Radiation, was used for instrument's calibration.

Optimal window was selected according to the highest FOM value achieved. The type of vial directly influences the Cherenkov background rate, so three types of vials (low diffusion polyethylene vials, high performance glass counting vials and low potassium borosilicate glass vilas of 20ml volume) were used in order to examine their potential effect on counting process.

Calibration curves were obtained by comparison of measured count rates of prepared solution of the certified standard and the activity added to water. The measurement efficiency  $\varepsilon$  was determined as the slope of the fitted line (linear fit with intercept set to 0).

Determined measured efficiencies, background count rates, average SQP values and detection limit  $DL$  for different types of vials are presented in Table 1.

The European Commission has established maximum permitted levels for the  $^{90}\text{Sr}$  contamination of water at  $125 \text{ Bq L}^{-1}$  (Euratom, 1989).  $^{90}\text{Sr}$  activity concentration in drinking waters collected from Serbia will be presented with annual effective dose for ingestion for adults calculated.

Table 1. Measured efficiencies, background count rates, average SQP values and detection limit  $DL$  for different types of vials

Type of vial	window (ch)	$\varepsilon$ [%]	$R_b$ [cps]	SQP (E)	DL [ $\text{Bq L}^{-1}$ ] (300 min)
Plastic	130-430	45.86(0.09)	0.008	323	
Glass	130-430	32.45(0.19)	0.021	302	
Low $^{40}\text{K}$ glass	130-430	32.24(0.14)	0.014	301	

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# Color quench correction for gross alpha/beta measurements in waters

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**Keywords:** Liquid Scintillation Counting, alpha/beta spectroscopy, color quench, PSA discriminator, Quantulus 1220™.  
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Liquid Scintillation Counting (LSC) is quite simple and relatively rapid method for simultaneous determination of gross alpha/beta activities with close to 100% counting efficiency for alpha and high energy beta emitters along with spectral information provided, which makes it ideal for natural radioactivity monitoring in drinking waters. Nevertheless quenching phenomenon appears to be the most problematic effect during detection by LSC techniques. The need for color quench correction have been recognized since natural water samples that require gross alpha/beta measurements can be significantly quenched (e.g. artesian well waters are not always transparent, having yellow color that probably origins from the presence of iron and its oxides impossible to bleach).

Two methods for color quench correction have been considered and evaluated in this paper, as a proposal for supplement to the ASTM method for gross  $\alpha/\beta$  determination. Measurements were performed by low-background liquid scintillation counter Quantulus 1220™, equipped with Pulse Shape Analysis (PSA) circuit that distinguishes  $\alpha/\beta$  events. Since optimization of alpha/beta spectra separation parameter is known to be strongly dependent on quench level, the first method was to readjust PSA setting to corresponding quench indicating parameter (SQP(E)) for each of the measured quenched samples. Alternatively, if all samples were counted on usual optimal PSA setting, count rates have been corrected afterwards according to previously determined quench calibration curve.

The following experiments were performed in order to verify PSA vs. SQP(E) correlation on one side, and count rate reduction curve dependent on quench level validity on the other, in case of color-quenched samples. For that purpose results of measured and corrected  $\alpha/\beta$  activities for some of the yellow water samples from artesian wells in Vojvodina region in Serbia, as well as few spiked and colored samples ( $^{241}\text{Am}/^{90}\text{Sr}$  of known activity in different ratios) have been compared and discussed.

Performed experiments also make contribution to investigations on the manner in which color quench affects optimal PSA parameter setting and  $\alpha/\beta$

interference factor and further on, their altogether influence on gross alpha/beta activity measurements.

Results of experiments indicate that method of application of counts reduction curve dependent on quench parameter gives activities in better agreement with their reference values.

## Acknowledgments

The authors acknowledge the financial support of the Ministry of Education, Science and Technological Development of Serbia, within the projects Nuclear Methods Investigations of Rare Processes and Cosmic No. 171002, Biosensing Technologies and Global System for Continuous Research and Integrated Management No. 43002.

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# Medical $^{131}\text{I}$ in River Water and Sediments

## Case study of Weser River in NW Germany

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### Introduction

City wastewater is known to be contaminated with considerable amounts of medically used radionuclides. Routinely the Bremen sewage plant releases effluent with  $^{131}\text{I}$  activity concentration, ranging between 35 and 1000 mBq/l (IMIS 1994-2014), into the water body of Weser River.

This work is focused on presenting the results of several ongoing sampling campaigns in the Weser River, for both river water and sediments in the period of summer 2014 - summer 2015 and studying the distribution of radioisotopes in the water-sediment system using medical  $^{131}\text{I}$  as readily available tracer.

### Study area

The sewage treatment plant is located about 8 km downstream of the city of Bremen in Northwest Germany (river 0 km). The selected sampling locations are in the river section in the vicinity of the plant, which is part of the tidal section of the Weser River. Additionally samples are taken upstream the Weir (-4.5 km) and at the sewage plant to ascertain the source of the  $^{131}\text{I}$  in the specific location.

### Sample treatment and analysis

Due to low  $^{131}\text{I}$  activity concentration in Weser River water, a chemical extraction method is applied in 50 L water samples in order to increase the sensitivity of the measurement. Detailed description of the procedure was given at the ICRER 2014 in Barcelona. (Souti M., et al., 2014)

The sediment samples are collected with a sediment grabber from the river bottom and from both banks of the river, measured in wet state and dried.

All samples are measured in high resolution, low background gamma spectroscopy by high purity germanium detectors of 50% relative efficiency. Spectra are evaluated using the Genie-2000 software by Canberra Industries.

### Results and discussion

During previous work on the topic from several group members over the period 2008-2012 a longitudinal  $^{131}\text{I}$  profile in Weser River was created, as shown in figure 1.

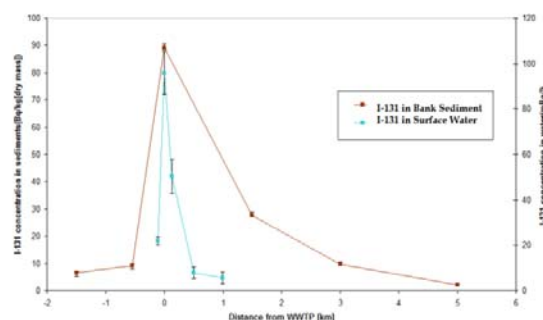


Fig. 1: Longitudinal  $^{131}\text{I}$  profile in Weser River (positive and negative values in the x-axis indicate down and upstream distances from the sewage plant). (Souti M., 2012)

Similar behaviour and dilution factors show the results from the recent sampling campaign on the river at November 2014, as indicated in table 1.

Table 1: Results from sampling campaign in Weser river-surface samples. River flow towards the coast.

Downstream distance from WWTP outlet (km)	WWTP activity concentration (mBq/kg)	Mass of sample (kg)	I-131 activity concentration (mBq/kg)	Dilution
0	327 $\pm$ 24	48.66	87.31 $\pm$ 11.98	3.74
0.5	327 $\pm$ 24	47.84	2.41 $\pm$ 0.76	135
1	327 $\pm$ 24	47.74	<1.38	>237

### Outlook

Over the period of the next few months, systematically water and sediments samples will be collected and analysed in order to determine the  $^{131}\text{I}$  distribution in the Weser River.

### Acknowledgments

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# Research of leaching of artificial radionuclides from soil of the «Balapan» testing site

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The «Atomic» lake was formed as a result of excavation explosion at «Balapan» testing site at the Semipalatinsk Test Site (STS). The heap of soil around the «Atomic» lake is a serious potential source of radioactive contamination of other environmental objects. The existing radiation situation can be aggravated because Shagan river is flowing through the «Atomic» lake and flows into Irtysh river. It can be cause of possible transfer of artificial radionuclides from «Atomic» lake to the other environmental objects (Aidarkhanov et. al, 2013).

This work aimed at research of possibility of artificial radionuclides transfer from soil into natural water by leaching. The main problems of research involved with the determination the degree of radionuclide leaching and study of factors, having influence on this process (dependence of degree of leaching on phase contact time and soil texture).

Samples of soil from the soil heap of the "Atomic" lake were taken as a research objects. They were sampled at the distance of 50 and 100 m from the coast line of the lake. Concentration of artificial radionuclides at these points differs by an order. Experiment on leaching were carried out under static conditions (Smirnova et al, 2009). Distilled water was used as a leaching solution (ratio of solid to liquid phase was 1:2). Aqueous extracts have been collecting in some definite time intervals (from 1 h to 100 days) for determining of artificial radionuclides concentrations. The analysis of soil was also performed after leaching. The degree of radionuclide leaching from the soil have been assessed based on obtained data.

According to the research results it was found a linear dependence the leaching degree of radionuclide  $^3\text{H}$  and  $^{90}\text{Sr}$  on the time. Concentration of  $^{239+240}\text{Pu}$  in water extracts have sharply increased during first 5 days, then it started to decrease and on the 60th day concentration of  $^{239+240}\text{Pu}$  in the water extract was below the detection limit of the technique used. Concentration of gamma-emitting radionuclides ( $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ ,  $^{152,154}\text{Eu}$ ) in the solution was below the detection limit of the used technique. It was

confirmed that transfer of radionuclides from soil of the «Atomic» lake into water is possible.

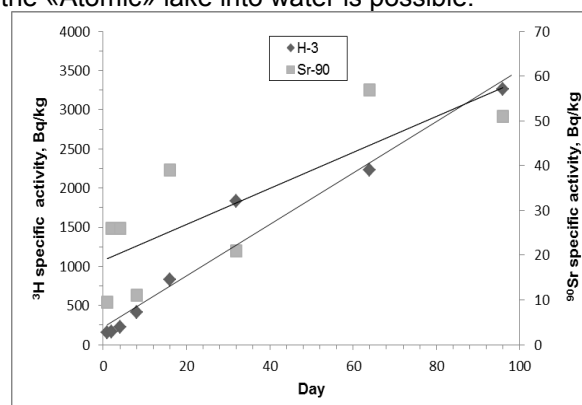


Figure 1 Concentration of  $^3\text{H}$ ,  $^{90}\text{Sr}$  in water extracts.

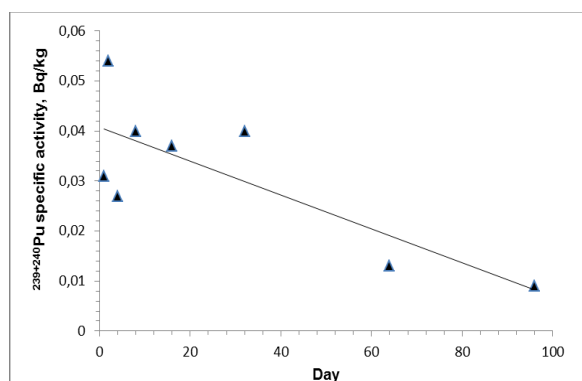


Figure 2 Concentration of  $^{239+240}\text{Pu}$  in water extracts.

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# First determination of Be-7 in fish from the South Adriatic: *Liza* species (*L. aurata*, *L. ramada*, *L. saliens*)

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Keywords: *Liza* species, South Adriatic, Be-7.  
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A previous research showed <sup>7</sup>Be activity concentration in the South Adriatic seawater less than 0.14 Bq L<sup>-1</sup>, but also relatively high concentrations in mussels – from 4.57 to 18.6 Bq kg<sup>-1</sup>, indicating that they accumulate this cosmogenic radionuclide in some way (Zizic *et al.*, 2001). As far as is known, <sup>7</sup>Be in other aquatic species from this marine environment has not been measured before this study, which is aimed to contribute to already established knowledge about radioactivity levels in different fish species. For example, from a previous research on <sup>137</sup>Cs and <sup>222</sup>Ra in the South Adriatic *Chelon labrosus* (Risso, 1826), Antovic and Antovic (2011) found a level up to 1.6 and 2.1 Bq kg<sup>-1</sup>, respectively (in whole individuals).

Among the Euro-Mediterranean mullet species occurring in the South Adriatic adjacent to the Coast of Montenegro, three are from the *Liza* genus: *Liza aurata* (Risso, 1810), *Liza saliens* (Risso, 1810) and *Liza ramada* (Risso, 1826), and they were considered for <sup>7</sup>Be activity, together with seawater and sample from the Sea floor (bottom sample).

Thirty-three *Liza* specimens were sampled by a trawl net in 2013 from the locality Solila – area of Tivat (Boka Kotorska Bay), and analyses included three whole individuals, eight muscles and two gastrointestinal systems of each *Liza* species. The samples were prepared and measured in a standard procedure by the ORTEC HPGe spectrometers, as well as seawater and bottom sample.

In seawater 0.05 Bq L<sup>-1</sup> of <sup>7</sup>Be was measured, in bottom sample (containing sand, mud and mussels) – 6.7 Bq kg<sup>-1</sup>, whilst in whole *L. aurata* and *L. saliens* – up to 12.3 and 12.1 Bq kg<sup>-1</sup>, respectively (in *L. ramada*: <6.37 Bq kg<sup>-1</sup>). Muscles showed lower level of <sup>7</sup>Be, but gastrointestinal systems – 29.9 (*L. aurata*), 34.9 (*L. ramada*) and 29.2 Bq kg<sup>-1</sup> (*L. saliens*).

Concentration factors for seawater to fish (CF<sub>1</sub>) and bottom to fish (CF<sub>2</sub>) were also evaluated using ratios of <sup>7</sup>Be activity concentrations (wet wt fish/seawater and wet wt fish/dry bottom sample, respectively). The CF<sub>1</sub>s to whole individuals showed an average equal or less than 147, 95.8 and 154 – for *L. aurata*, *L. ramada* and *L. saliens*, respectively, as CF<sub>2</sub>s – 1.09, 0.71 and 1.15, respectively. Both CFs were found to be significantly higher for gastrointestinal systems than for whole fish. In regard to muscles (CFs shown in Figure 1), CF<sub>1</sub>s minimum, maximum, average and standard deviation were – 108, 228, 163 and 43.6, respectively (*L. aurata*), 98.4, 286, 148 and 60.7, respectively (*L. ramada*), 77.6, 141, 115 and 23.2, respectively (*L. saliens*); CF<sub>2</sub>s – 0.8, 1.7, 1.22 and 0.32, respectively (*L. aurata*), 0.73, 2.13, 1.11 and 0.45, respectively (*L. ramada*), 0.58, 1.05, 0.86 and 0.17, respectively (*L. saliens*).

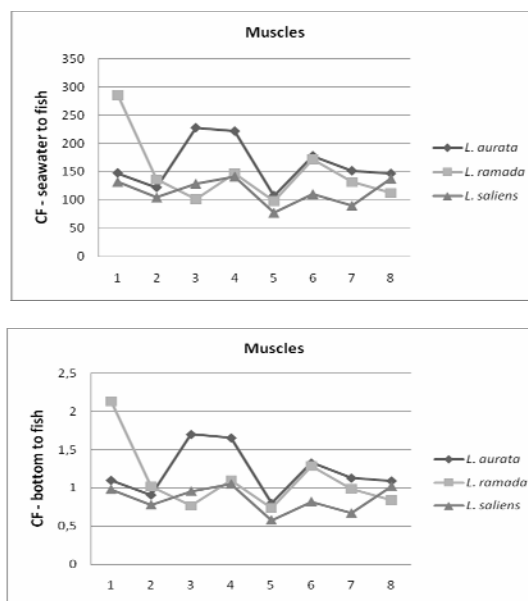


Figure 1. Concentration factors for <sup>7</sup>Be – from seawater and bottom to fish muscles.

Comparing these results with those obtained for fish fauna from the Yenisei River (Siberia, Russia), where 17.7 Bq kg<sup>-1</sup> was observed in cod (Zotina *et al.*, 2011), it can be concluded that, despite the fact that in whole fish and muscles of the *Liza* species detected activities were lower, their gastrointestinal systems showed somewhat higher <sup>7</sup>Be activity. Such levels might be explained by the *Liza* feeding. These species, as well as other mullets, feed on detritus, mud and organics from the Sea floor.

## Acknowledgments

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# The effectiveness of wastewater treatment in Nuclear Medicine Performance data and radioecological considerations

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Keywords: radioiodine therapy, wastewater treatment, radioecology.  
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The administration of radiopharmaceuticals in Nuclear Medicine leads to a radiation exposure in several respects: patients, staff members and members of the public. One important radioecological aspect is that the radionuclides are excreted by the patient after undergoing diagnostic and therapeutic procedures and without further consideration they would be released to the environment.

With a total of 50000 radioiodine therapies and two million diagnostic procedures in Germany alone this could account for a significant elevation of the respective radionuclide concentrations in the environment. In Germany it is therefore mandatory to store and document the radioactive inventory and wastewater is only released to the public sewage when the respective activity concentrations fall below the exemption level.

This work presents exemplarily the design and operation details of the wastewater plant of the University Hospital of Cologne.

The wastewater treatment is based on a decontamination plant ("cooling plant") and consists of a closed tank-stack system where the liquid waste is stored and measured automatically by in-tank scintillation counters while decaying as described by Westmeier (2012). All measurements are recorded.

The cooling plant consists of 19 tanks divided into five subgroups (therapy ward (13), diagnostic procedures (2), laboratories (4)). The total capacity of the plant is 305 m<sup>3</sup>.

The whole radioactive inventory depends on the activities administered to the patient and a significant build-up over the time is only seen for longer-lived nuclides like <sup>131</sup>I which are used for therapeutic purposes.

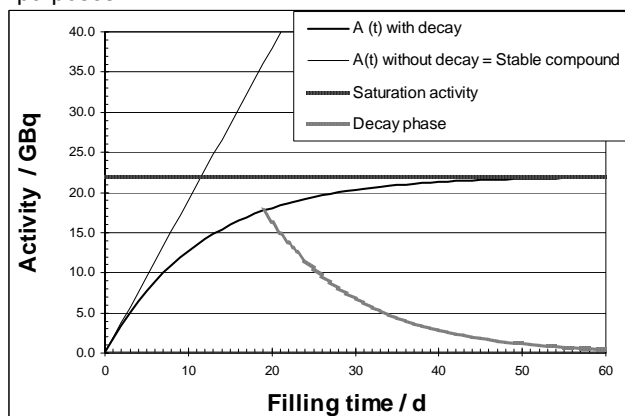


Figure 1. Radioactive inventory (<sup>131</sup>I) of a wastewater plant for a nuclear medicine therapy ward

The most important diagnostic radionuclides with half-lives between 20 min and 2,7 d (<sup>11</sup>C, <sup>68</sup>Ga, <sup>99m</sup>Tc, <sup>123</sup>I) decay almost entirely within the filling period of a tank.

The inflow of <sup>131</sup>I from the therapy ward can be calculated precisely by various means as described by Wellner et al. (1993). When the activity concentration after complete filling of one tank is determined by measurements, the whole radioactive inventory can be calculated (Fig. 1).

A steady state activity of 22 GBq could be derived for the years 2003 - 2006. This value corresponds to an annually administered activity of roughly 1 TBq (~1000 radioiodine therapies). The annual discharges from the therapy ward were found between 300 kBq - 7 MBq, hence only a few ppm of the annually administered activity were discharged. Since 2007 the inflow was reduced leading to longer filling periods. Annual discharges therefore decreased significantly so that since 2011 only a few kBq are released (Tab.1).

Table 1. Actual releases of radioiodine from the wastewater plant

Year of recording	Section A		Section B	
	c <sub>A</sub> (Bq / l)	V (m <sup>3</sup> )	c <sub>A</sub> (Bq / l)	V (m <sup>3</sup> )
2011	0.064	323	0,07	24
2012	0.004	209	0.04	12
2013	0.008	247	0.20	12
2014	0.001	228	0	0

The cooling plant as described in this contribution reduces the amount of radioactivity released into the environment effectively by six to eight orders of magnitude (2002 - 2014) when compared to the administered activity. This exceeds even the high effectiveness of decay systems as discussed by Goddard (1999) by far. The discharge to the public sewage system can be minimized to a large extent predominantly by reducing the inflow into the plant.

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# Organically Bound Tritium activities within French continental watersheds

## Preliminary results from areas most impacted by past nuclear fallout

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**Keywords:** Tritium, Organic matter, Sediments, Coastal Hydrosystems  
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Naturally occurring in the atmosphere due to their cosmogenic origin, Tritium ( $^3\text{H}$ ) and Carbon 14 ( $^{14}\text{C}$ ) has been massively released in the environment by atmospheric nuclear weapons testing from 1945 to 1980 leading to activities in rains respectively more than one hundred times and two times higher than natural levels (UNSCEAR, 2000) and (Levin and Hesshaimer, 2000) generating an increase in the  $^{14}\text{C}/^{12}\text{C}$  ratio in the atmosphere at the global scale.

These isotopes are currently the most released by nuclear facilities and unlike main trace elements,  $^{14}\text{C}$  and  $^3\text{H}$  largely integrates into both the water and carbon cycles.

Even though naturally encountered in gaseous forms (HT, CH<sub>3</sub>T), tritium is essentially present as tritiated water (HTO) also called the “free” form fostering its integration into the various biological components. Thus, it can be bound to the organic matter through two main specific ways. According to the kind of bind, Exchangeable or Non-Exchangeable Organically Bound Tritium (E-OBT and NE-OBT, respectively) are expected to be encountered within the biosphere. This close integration within the carbon cycle is governed by biological processes, mainly photosynthesis but also through metabolic reactions independent of bright conditions (Diabaté and Strack, 1997). The further behaviour of these organically bound forms is then thought to closely refers to the biodegradation cycle of organic compound (Eyrolle-Boyer et al., 2014).

A recent assessment of OBT activities data into different aquatic components (water, sediments or aquatic plants and fishes) from areas located outside from the direct influence of tritiated releases from nuclear industries indicate an important range of variability of OBT activities in a same matrix referring to the free form referential values registered in freshwaters or rain fall.

These disequilibrium between the bound and free forms are assumed to be fully related to the organic matter origin and quality. Indeed, deferred transfert of allogenic detrital organic matter initially contaminated by global atmospheric fallout from nuclear weapons testing's are thought to be responsible for most of the currently observed disequilibrium's.

This work first aims to underline the original methodology adopted to select sampling areas corresponding to theoretical global fallout persistence areas in order to test our hypotheses. Preliminary results

on OBT and  $^{14}\text{C}$  activities measured in the various components of the focussed Areas will be discussed.

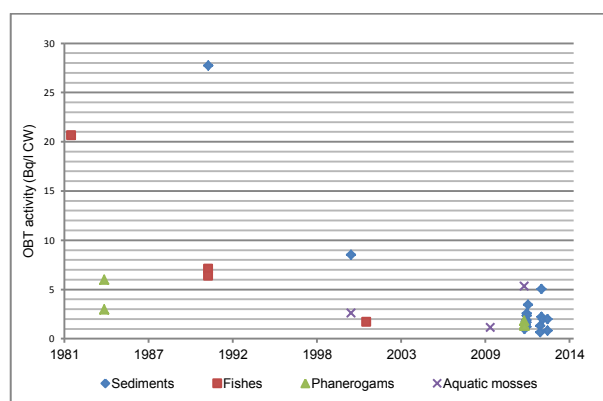


Figure 1. Relation between OBT activities in several matrices of different french coastal hydrosystems and  $^3\text{H}$  activities in rainwater at Vienne (AUT)

### Acknowledgments

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# A Study of Artificial Radionuclides in Lake Sediments

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Keywords: Lake, sediment, <sup>134</sup>Cs, <sup>137</sup>Cs, <sup>131</sup>I, global fallout

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After Fukushima Daiichi nuclear power plant (NPP) accident on March 11<sup>th</sup>, 2011, various studies have been carried out in the terrestrial environment and marine environment for monitoring artificial radionuclides. Nevertheless, the monitoring in the public waters such as river and lake wasn't conducted much. The Ministry of Environment established a law for monitoring the artificial radionuclides (Cs-134, Cs-137 and I-131) in public waters in 2014.

The objectives of this study are to access the contamination of artificial radionuclides and to improve the monitoring systems in lake sediment, water and fish from Lake Euam, Chuncheon in South Korea.

sediment cores (4.4 cm in diameter) were collected from 4 sites on July 29-30<sup>th</sup>, 2014. After sampling, the samples were immediately sliced about 1 cm interval and put into plastic pack. After carried to lab, sliced samples were freeze-dried and sieved through a 2.0 mm pore size. Samples were transferred to a standardized container for a well-type HPGe detector (Ortec) with a relative efficiency of 90%.

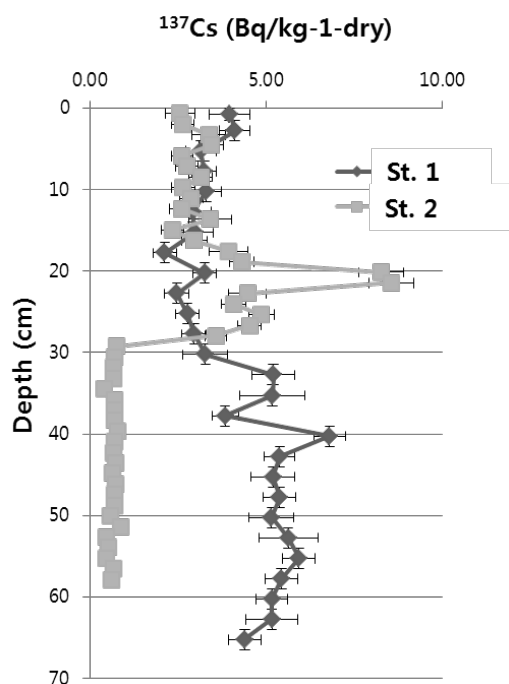


Figure 1. The profiles of <sup>137</sup>Cs concentration in the sediment at St. 1 and 2

The concentration of <sup>137</sup>Cs at St. 1 & 2 is presented in Fig. 1.

The <sup>137</sup>Cs concentration at St. 1 is ranged from  $2.1 \pm 0.3$  Bqkg<sup>-1</sup>-dry to  $6.8 \pm 0.5$  Bqkg<sup>-1</sup>-dry. The concentration of <sup>137</sup>Cs increases gradually with depth. The <sup>137</sup>Cs concentration at St. 2 is ranged from  $2.6 \pm 0.4$  Bqkg<sup>-1</sup>-dry to  $8.6 \pm 0.6$  Bqkg<sup>-1</sup>-dry. From surface to 17 cm, the <sup>137</sup>Cs concentration is relatively constant, then at depth 20~22 cm, its concentration increases sharply with showing a highest concentration. Below 29 cm, the <sup>137</sup>Cs concentration represents MDA values. The concentration of <sup>137</sup>Cs found in the lake sediment is comparable to that of <sup>137</sup>Cs in soil (Lee et al., 2013).

The concentration of <sup>137</sup>Cs below 29 cm depth at St. 2 represents Minimum Detectable Activity (MDA) values ( $< 0.89$  Bqkg<sup>-1</sup>-dry).

On the other hand, at St. 1 and 2, the <sup>134</sup>Cs and <sup>131</sup>I concentrations are reported less than MDA value i.e., for <sup>134</sup>Cs  $< 1.06$  Bqkg<sup>-1</sup>-dry and for <sup>131</sup>I  $< 0.86$  Bqkg<sup>-1</sup>-dry.

<sup>134</sup>Cs/<sup>137</sup>Cs ratio released Fukushima Daiichi NPP accident is ca.1 (the present ratio is ca. 0.35 by decay correction). At the St. 1 & 2, the <sup>134</sup>Cs concentration represents MDA value, and it indicates that the Cs in sediment is originated from global fallout, not Fukushima Daiichi NPP accident.

In the northern hemisphere, the highest deposition of global fallout was found in 1963. Consequently, sediment layer with the highest <sup>137</sup>Cs concentration at St. 2 is supposed to be 1963.

Additional analysis for St. 3 & 4 including Pu isotopes is ongoing and will be reported later.

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# ACTIVITY CONCENTRATIONS OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN FELINE DRY FOOD

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Keywords: animal food, natural radionuclides, artificial radionuclides, gamma spectrometry.  
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Natural radiation exposure is an inherent condition to all living species, once radionuclides from the  $^{238}\text{U}$  and  $^{232}\text{Th}$  chain can nearly be found in all places. Information on radionuclides concentration and exposure levels, from natural and anthropogenic sources are absolutely necessary to investigate the possible effects that ionizing radiation can induce. These can be very different depending on the organism considered and the exposure pathway. In recent decades, the exposure of non-human species to ionizing radiation has been specially considered and investigated (ICRP, 2014) by a vast number of scientists and organizations, once they differ widely from the exposure of human beings.

Brazil holds the second largest cat and dog population in the world, consuming over 2 million tons of feed every year. The country also stands out for its production of pet food that produced 2.4 million of tons of feed in 2014, representing the world's second largest industry. A novel study regarding the radionuclide content in different dog and cat food is being developed since 2013 and preliminary results have been presented by Cavalcante, F. and Pecequilo (2014), for selected dry dog food. The present study presents an evaluation of the radionuclide and radioactivity content of different brands of dry cat food, commonly found in local markets in the city of Sao Paulo, Brazil.

Thirteen different samples were crushed into powder and kiln dried before tightly sealed in 100 mL HDPE flasks, with a plan screw cap and bubble spigot. These samples, after resting for 30 days to ensure secular equilibrium, were placed in an extended range coaxial germanium detector (Canberra XiRa GX4020 detector) for 150 ks and the acquired spectra were analyzed with the InterWinner 6.0 software (InterWinner, 2004). The natural radionuclides considered were  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , the anthropogenic radionuclides investigated were  $^{60}\text{Co}$ ,  $^{131}\text{I}$ ,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ .

The results for the considered artificial radionuclides have shown activity concentration values below the detector's MDA (Minimum Detectable Activity), as in Table 1.

Table 1. MDA (Minimum Detectable Activity) range values for the considered artificial radionuclides.

	$^{60}\text{Co}$	$^{131}\text{I}$	$^{137}\text{Cs}$	$^{134}\text{Cs}$
MDA (Bq/kg)	0.8 – 1.0	0.7 – 1.1	0.8 – 1.0	0.8 – 1.1

The concentrations of natural radionuclides ranged from  $1.12 \pm 0.29$  Bq/kg to  $3.77 \pm 0.36$  Bq/kg for  $^{226}\text{Ra}$ ; from  $1.48 \pm 0.40$  Bq/kg to  $6.27 \pm 0.78$  Bq/kg for  $^{232}\text{Th}$  and from  $216.8 \pm 11.2$  Bq/kg to  $361.7 \pm 16.8$  Bq/kg for  $^{40}\text{K}$ , as shown in Figure 1.

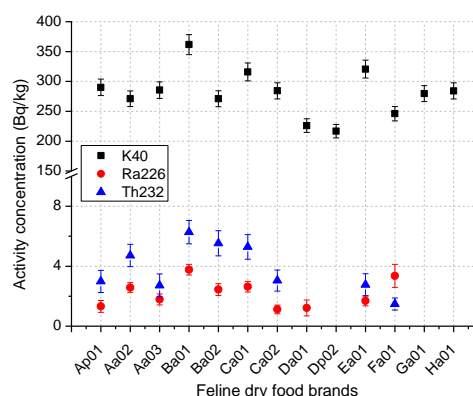


Figure 1.  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  activity concentration values for 13 different samples of dry cat food. Producers brands are labelled from A to H, (a) meaning adults and (p) puppy cats.

The results suggest that the samples evaluated have no contamination of artificial radionuclides and the natural radionuclides concentration will not contribute to significant absorbed dose by their ingestion. Therefore, the authors conclude that these studied brands carry no radiological risk for the animals ingesting them.

## Acknowledgments

F. Cavalcante would like to thank CAPES for the scholarship grant.

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# RADIOANALYTICAL ASSESSMENT OF SEDIMENTATION RATES IN THE CARAVELAS ESTUARY (BAHIA, BRAZIL) USING UNSUPPORTED $^{210}\text{Pb}$ AND $^{137}\text{Cs}$ MODELING.

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Keywords:  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ , Models, Sedimentation Rate, Caravelas Estuary

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The region of Caravelas is one of the oldest urbanized areas of northeast Brazil. The region has great ecological significance as 15 km into the sea from the Caravelas Estuary is located the Abrolhos Archipelago, the richest and most extensive reef complex (6,000 km<sup>2</sup>) in the South Atlantic Ocean (Travassos et al., 2006).

There is a lack of knowledge regarding quantitative measures of sedimentation, such as sedimentation rates for this area of the Caravelas Estuary. Therefore, this study aimed to evaluate the recent (time range of approximately 150 years) sedimentation rates in three sediment profiles collected along the Caravelas Estuary.

Vertical sediment cores were manually collected in different regions of the estuary. These cores were sliced in to 2 cm layers and transferred to cylindrical polyethylene containers proper for gamma spectrometry analysis in low-background hyperpure Ge gamma spectrometer (EGG&ORTEC, model GMX25190P).

Three models were used to assess the sedimentation rates in Caravelas Estuary. The CIC (Constant Initial Concentration), the CRS (Constant Rate of Supply) and the *fallout*  $^{137}\text{Cs}$  (Figures, 1, 2 and 3).

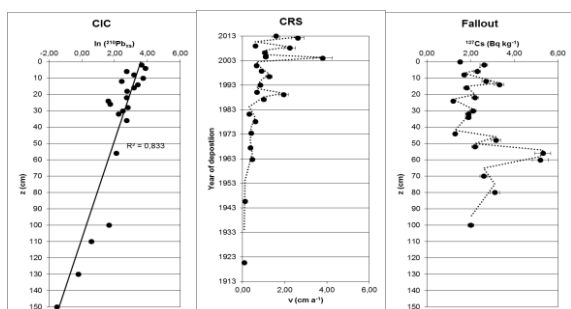


Figure 1. Models of sedimentation for T#2 of the Caravelas Estuary.

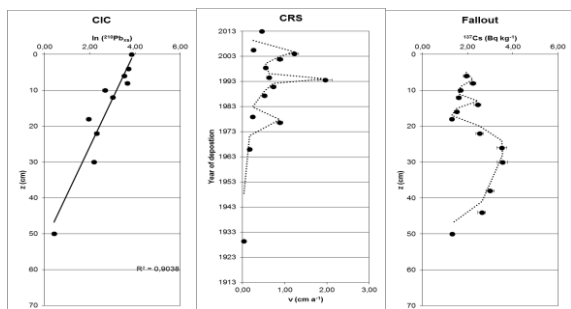


Figure 2. Models of sedimentation for T#5 of the Caravelas Estuary.

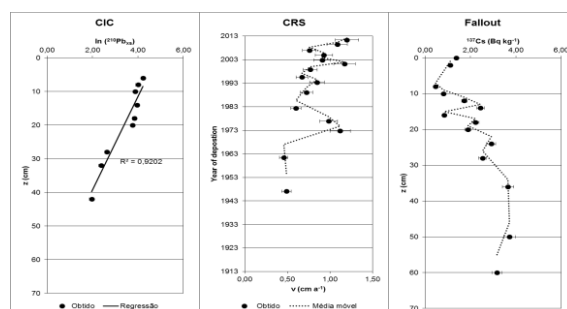


Figure 3. Models of sedimentation for T#8 of the Caravelas Estuary.

From these results, it was possible to establish the sedimentation rate for the cores collected in the Caravelas Estuary, the obtained results are expressed in Table 1.

Core	Sedimentation rate (cm year <sup>-1</sup> )		
	CIC	CRS	<i>fallout</i> $^{137}\text{Cs}$
T#2	1,13 ± 0,13	1,07 ± 0,13	1,12 ± 0,08
T#5	0,37 ± 0,04	0,65 ± 0,06	0,56 ± 0,04
T#8	0,66 ± 0,07	0,85 ± 0,09	0,72 ± 0,05

Table 1. Sedimentation rates, using the model CIC, CRS and  $^{137}\text{Cs}$  *fallout* in sediment cores of the Caravelas Estuary.

The difference in the sedimentation rates between the sample cores could be related to hydrodynamics processes. The inner region, represented by T#2 have higher sedimentation rates due to the decrease of intensity of the currents, showing the importance e influence of the mangrove system. Meanwhile, the outer region expressed by T#5 and T#8, tend to be dominated by intense water flux which hinders the deposition of sediments.

## Acknowledgments

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# Uranium in surface water at a Naturally Occurring Radioactive Material area in Brazil

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Keywords: NORM area, environmental radioprotection, radioecology, natural radionuclides.

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The region of Santa Quitéria, Ceará, Brazil, is rich in phosphate associated with uranium, being an area of Naturally Occurring Radioactive Material - NORM (Pereira, Kelecom & Pereira, 2014).

The concentration of activity (CA) of <sup>238</sup>U in surface water of the region has been determined, and the distribution of uranium in the soluble phase (the one that passes through a 0,45 µm filter) and the particulate phase (retained on a 0,45 µm filter) has been assessed.

Six collecting points were chosen (01SQ a 06SQ), distributed spatially in the surrounding area of the deposit; water samples were collected between October 2009 and December 2011, for a total of 300 samples.

The data were submitted to an analysis of variance (ANOVA) to identify differences between the points of collection and between the analyzed fractions (Ceteno, 1999).

When differences appeared, data were grouped using the "Tukey" test. Then a Student "t" test was carried out between fractions of the same point. Finally, a principal component analysis (PCA) was performed, in order to group data (Vallentin, 2000).

The means per point and fraction are shown in Figure 1. The ANOVA, among points/fractions indicated that at least one set of point/fraction had CA different from the others ("F" value of 221, P <0.01).

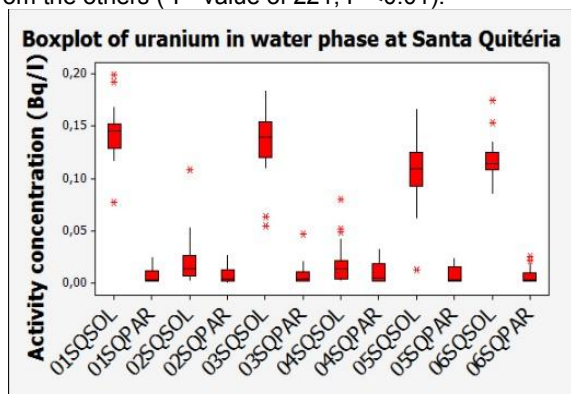


Figure 1 – Means concentrations of activity of <sup>238</sup>U.

The PCA (Figure 2) differentiated the fractions and showed that the particulate fraction is more homogeneous than the soluble one, corroborating the "Tukey" test.

The "t" test, applied on CA of <sup>238</sup>U between fractions by point, demonstrated that the CA of soluble fractions are considered statistically higher to those of particulate fractions, except for point 04SQ where they were considered statistically identical.

The "Tukey" test showed that the soluble fraction had higher concentrations of activity than the particulate one. It also showed that the soluble fraction is

heterogeneous, forming 4 subgroups (A-D) while the particulate fraction is homogeneous with only one group, (D) as can be seen in Table 1.

Table 1 – "Tukey" test at the collecting points/fraction.

Point/fraction	N	Mean (Bq.l <sup>-1</sup> )	Group <sup>1</sup>
01SQsoluble	25	0,144	A
03SQsoluble	25	0,136	A B
06SQsoluble	25	0,118	B C
05SQsoluble	25	0,103	C
02SQsoluble	25	0,021	D
04SQsoluble	25	0,018	D
04SQparticulate	25	0,008	D
03SQparticulate	25	0,008	D
02SQparticulate	25	0,008	D
05SQparticulate	25	0,008	D
06SQparticulate	25	0,007	D
01SQparticulate	25	0,006	D

<sup>1</sup>Items under the same letter have identical means.

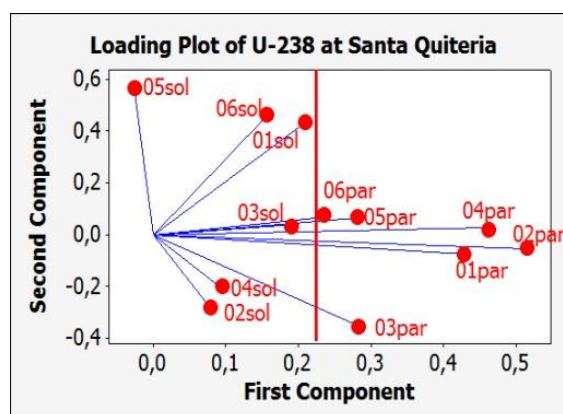


Figure 2 – PCA between points/fractions.

The analysis of data indicates that there is a geographical difference between the CA of <sup>238</sup>U and also an unequal distribution of <sup>238</sup>U between the fractions. Thus, the soluble fraction has CA of <sup>238</sup>U superior to the particulate fraction and the differences between the points are due to the soluble fraction.

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# Total alpha-beta radioactivity of bottled drinking water in Turkey

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Water that is the keystone of life carries physical and chemical properties due to the several parameters such as the path of the underground, the region's natural resources, environmental factors and their interaction time in the process of reaching the earth. The element of water and minerals in the water gives the form the chemical and radioactive properties. Radionuclide are located within the water will cause its own specific natural activity. Chemical, radiological and bacteriological analyzes are applied to the water for a healthy life and control of the allowable limits.

Accordingly, the water is performed analyzes on the presence of radioactive elements. These analyzes include radon in water, uranium, methods such as detecting the amount of radioactive elements such as radium and thorium. First method is the analysis of total radioactivity in the alpha-beta often being preferred for the preliminary assessment and rapid assessment because of these methods are extremely expensive.

This study is aimed to measuring the total alpha-beta radioactivity of drinking water that consist commercially available which is consumed in Turkey and to make an evaluation of performed according to standard limit. Consequently, drinking water samples properties which are physical, chemical, biological, radiological features are defined using by total alpha-beta radioactivity methods.

**Keywords:** *Drinking water, Natural radioactivity, Total alpha-beta radioactivity, Low-level radioactivity, Bottled water.*

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# Determination of the radionuclide soil-to-plant transfer factors - laboratory methodology

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**Keywords:** transfer factor, methodology, plants, soil.

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In the frame of the research programme the methodology of determination of radionuclide transfer factors from soil to plants was developed.

The aim of the methodology was to define a clear and reproducible laboratory method of the soil-to-plant transfer factor determination. The particular steps and techniques are based on the IAEA documents TECDOC-1616 and TECDOC-1497, and meet the requirements of the gamma spectrometry laboratory of the National Radiation Protection Institute (SÚRO) in the Czech Republic.

The method of transfer factor determination was developed initially only for caesium radioisotopes because  $^{137}\text{Cs}$  is one of the most significant radionuclides, which can contaminate large areas for long time after serious nuclear power plant accidents. Even though the methodology deals with the caesium isotopes, it can be easily extended to other radionuclides, the content of which could be easily measured by low background gamma spectrometry using HPGe detectors. For example  $^{85}\text{Sr}$  as a test radionuclide for  $^{90}\text{Sr}$ , the next important radionuclide which can contaminate environment in the case of NPP accident, is suitable.

For the purpose of the methodology the transfer factor was defined as the ratio of the activity concentration of plants and the activity concentration of soil in correspondence with the IAEA definition

$$TF = \frac{A_{mP}}{A_{mS}}$$

where  $A_{mP}$  is the mass activity of the dried plant and  $A_{mS}$  is the mass activity of the dried soil.

The method was tested and validated on corn cultivated from grains seeded to homogeneously  $^{134}\text{Cs}$  contaminated soil. The procedure of the transfer factor determination consisted of the soil contamination, homogeneity check of the contamination, plant seeding, growing, harvesting and drying the above-ground part of the plants and its measurement using low background gamma spectrometry to determine the  $^{134}\text{Cs}$  activity concentration.

According to the methodology, the plants were cultivated under the accurately specified condition in the cultivation box Q-Cell Vitrum in plastic containers or pots preventing the radionuclide leaks. The cultivation box keeps and controls the temperature, lighting and other settings, which are chosen with respect to needs of the particular plant species, simulated grow phase and season.

## Acknowledgments

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# Dating of a German riverine lake sediment using Pb-210, Cs-137 and Be-7.

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Keywords: Gamma-spectrometry, River sediment, Pb-210, Be-7, Cs-137 Chronology.

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Sediment chronology using CF-CS (Robbins et al, 1978) and CRS models (Appleby & Oldfield et al, 1978) is a method that have been used during the last half century for recent sediments.

Both models use certain assumptions (CF-CS: Constant Flux and Constant Sedimentation) and (CRS: Constant rate of <sup>210</sup>Pb<sub>xs</sub> supply), they have been tested in a great number of cores where presumably these conditions can be valid.

The Lower Havel is a riverine lake situated west of Berlin. It has a maximum depth of 11m and an average depth of 5.1m. The hydraulic residence time of the water is 18d on average due to the Havel river flowing through this lake (Grüneberg, et al. 2014).

In this work, a 32cm long core extracted in Oct.2014 was investigated. We attempted to use both classical models in a riverine lake environment where upper mentioned assumptions might be difficult assumed.

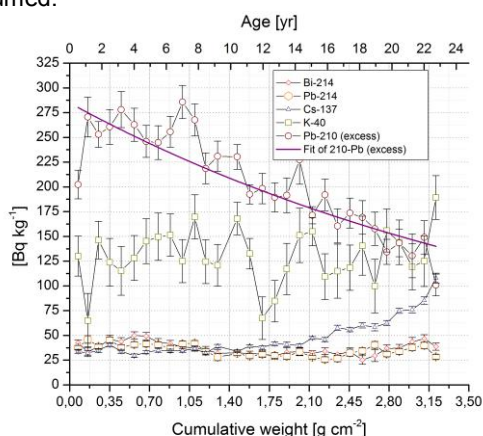


Figure 1. Depth distribution of the excess <sup>210</sup>Pb for lake Lower Havel and the exponential function fit (CF-CS chronology and calculating the missing inventory in the CRS model), together with activity concentration of <sup>214</sup>Bi, <sup>214</sup>Pb, <sup>137</sup>Cs and <sup>40</sup>K. Age in the x-axis was determined using the CF-CS model.

Results from the gamma-spectrometric determinations are presented in Fig. 1. Excess <sup>210</sup>Pb was obtained by the subtraction of <sup>210</sup>Pb supported (derived from <sup>214</sup>Pb assuming secular equilibrium between <sup>214</sup>Pb and <sup>226</sup>Ra) from the total. <sup>7</sup>Be activity of 31.02±13.48 Bq·kg<sup>-1</sup> in the uppermost section indicates recent sedimentation (age <1 year) and therefore the sampling date (10/2014) can be used as a reference date for the

core top. Mean calculated sedimentation rates can be found in Tab 1.

Table 1. Average mass sedimentation rates  $\bar{R}_s$  (g·cm<sup>-2</sup>·yr<sup>-1</sup>) for both models.

CF-CS	CRS
0.141±0.016	0.144±0.003

The presented results indicate a good agreement between the sedimentation rates derived by both models (Fig.2). The <sup>137</sup>Cs profile is in accordance with the <sup>210</sup>Pb chronology and the Chernobyl peak would be expected at 29 yr (equivalent depth ~39.5cm). On the other hand, a signal of <sup>7</sup>Be provides a time marker at the top of the core.

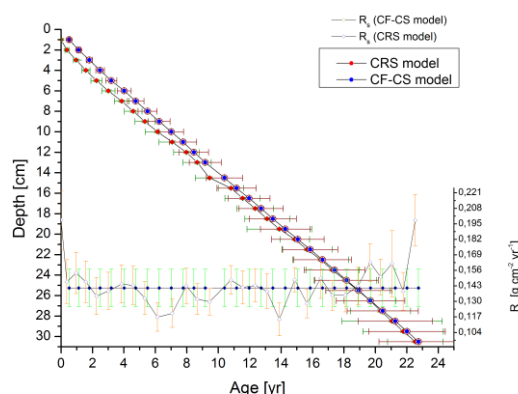


Figure 2. Chronology of sediments from lake Lower Havel. Comparison of the depth vs. age using models CF-CS (blue points) and CRS (red points). Sedimentation rates are plotted for both models.

## Acknowledgments

This work was supported by the Leibniz-Institute of Freshwater Ecology and Inland Fisheries (IGB). We want to thank Matthias Rothe for providing the sediment core.

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### Variation of sedimentation rate in the semi-enclosed bay determined by $^{137}\text{Cs}$ distribution in sediment (Kaštela Bay, Croatia)

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**Keywords:** sedimentation rate,  $^{137}\text{Cs}$ , Kaštela Bay, Adriatic Sea  
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Kaštela Bay coastal area is one of the most urbanized and most densely populated areas in Croatia and one of the most industrialized areas as well. It comprises the city of Split, Kaštela, Solin and Trogir towns, representing the largest urban agglomeration on the east Adriatic coast. The past and present industrial activities include chemical factory "Adriavinil", cementworks, ironwork and galvanization facility, all located in the east part of the area. Agricultural activities are more significant in the west part of the area. Large amounts of the untreated sewage waste waters, runoff and industrial waters were discharged into the Bay for decades and TENORM was also deposited in the Bay (Ujević *et al* (2000), Margeta (2002)) influencing sediment quality in the Bay (Orescanin *et al* (2005)). These discharges represented a significant anthropogenic source of particulate matter to be deposited in the Bay. The aim of the study was to determine influence of the anthropogenic activities on the sedimentation rates in the Bay considering a significant anthropogenic source of sedimentation material and spatially concentrated industrial activity and population density.

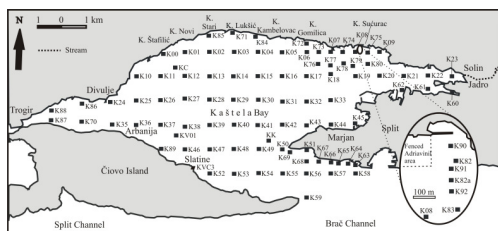


Figure 1. Sampling stations in the Kaštela Bay

Samples were taken at 95 sampling stations in a regular 1×1 km grid and in a 500×500 m grid around the "Adriavini" factory and along the profile next to the factory (Figure 1). Samples were taken by a gravity corer or by autonomous diving up to a 50 cm depth. Sediment cores were sliced into 5 cm and 10 cm segments. Sediment was dried at 105°C and ground. A peak at 661.7 keV was used to measure <sup>137</sup>Cs massic activity by gamma-spectrometry using a HPGe detector coupled with an 8192-channel multichannel analyzer (Canberra Industries). Sedimentation rate was calculated by dividing a depth of the marker peak (in cm) by the age of the sediment at the time of sampling (in years).

General increase of sedimentation rate in the whole Bay since 1954 and forming of localized

areas of markedly increased sedimentation rates in the east part of the Bay were found (Figure 2). Depending on the material input, sedimentation conditions, and bottom topography, localized areas vary in position.

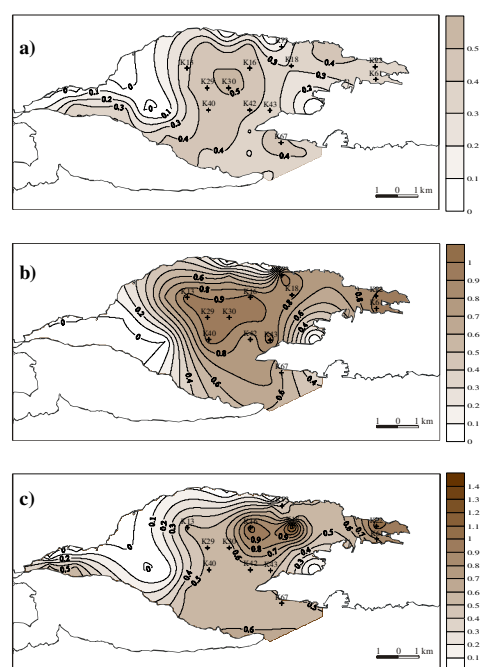


Figure 2. Sedimentation rates (in cm/yr) in the Kaštela Bay for three periods; **a)** 1954-2005; **b)** 1963-2005 (2006); **c)** 1986-2005 (2006); Reference stations are marked for comparison of rates from different periods.

## Acknowledgments

This work was supported by the Ministry of Science, Education, and Sports of the Republic of Croatia and by the Kaštela town.

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# Bioaccumulation of radiocesium in *Isochrysis galbana* and *Phaeodactylum tricornutum* and its assimilation in manila clam *Ruditapes philippinarum*

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In the current study phytoplankton species *Isochrysis galbana* and *Phaeodactylum tricornutum* exposed to dissolved  $^{134}\text{CsCl}$  ( $8 \times 10^4 \text{ Bq l}^{-1}$ ) in order to determine bioaccumulation kinetics of radiocesium. Bioaccumulation of radiocesium in *I. galbana* and *P. tricornutum* was followed during 20 and 7 days, respectively. Afterwards, manila clam *Ruditapes philippinarum* fed by radiolabelled phytoplankton. Following the dietary exposure the depuration of  $^{134}\text{Cs}$  was followed during 22 days in the clam.

The monocultures of *I. galbana* and *P. tricornutum* were growth in sterile f/2 medium. Medium was spiked with the  $^{134}\text{CsCl}$  ( $8 \times 10^4 \text{ Bq l}^{-1}$ ). The temperature of the culture media was  $21 \pm 1^\circ\text{C}$ . The uptake kinetics of  $^{134}\text{Cs}$  was followed 20 and 7 days in *I. galbana* and *P. tricornutum*, respectively. Plankton cultures were filtered with  $0.45 \mu\text{m}$  filter paper. Filtered plankton and seawater were counted in each counting day by using HPGe detector connected to a multi-channel analyser. Dry weight concentration factor was calculated for each counting day (Fig. 1).

The depuration kinetics of  $^{134}\text{Cs}$  was followed in the clam. A total of 20 clams acclimated to laboratory conditions for 20 days prior to the experiment. Sixteen of the clams were placed into separate glass containers. Thirty ml of radiolabeled phytoplankton culture was centrifuged and the pellet was put into the each glass containers. Each clam was fed with radiolabeled phytoplankton for one hour (pulse-chase feeding method; Warnau et al., 1996). After feeding period,  $^{134}\text{Cs}$  activity was measured in the clams using HPGe detector connected to a multi-channel analyser. Then clams were placed in uncontaminated seawater conditions (20-l PVC aquarium under flowing seawater  $1 \text{ l h}^{-1}$ , temperature:  $17 \pm 1^\circ\text{C}$ , salinity: 22 ‰). The depuration kinetics of  $^{134}\text{Cs}$  in clams was followed during 22 days.

The whole body depuration kinetics of  $^{134}\text{Cs}$  from radiolabelled phytoplankton was described by a two-component exponential model (Whicker and Schultz, 1982) (Fig 3).

Table 1. *Ruditapes philippinarum*. Depuration parameters:  $A_{0S}$  and  $A_{0I}$ : short and long lived exponential components activity (%), respectively;  $T_{b1/2}$ : biological half-life; ( $A_t = A_{0S} e^{-k_{tes} t} + A_{0I} e^{-k_{tel} t}$ ); ASE: asymptotic standard error)

Experiment	$A_{0S} \pm \text{ASE}$	$T_{b1/2S} \pm \text{ASE}$	$A_{0I} \pm \text{ASE}$	$T_{b1/2I} \pm \text{ASE}$
<i>I. galbana</i>	$93.7 \pm 12.3$	$1.5 \pm 1.0$	$6.3 \pm 0.9$	$16.8 \pm 0.8$
<i>P. tricornutum</i>	$84.9 \pm 9.8$	$2.5 \pm 0.8$	$15.1 \pm 2.0$	$58.5 \pm 2.6$

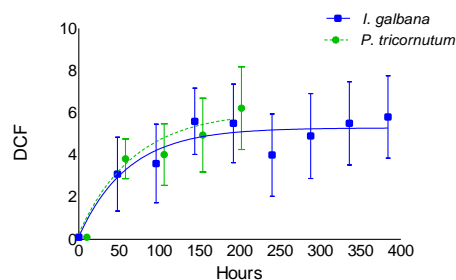


Fig. 1. Uptake kinetics of  $^{134}\text{Cs}$  in *I. galbana* and *P. tricornutum*

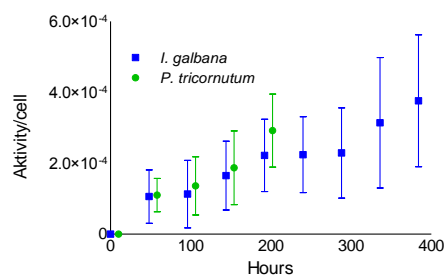


Fig. 2. Uptake kinetics of  $^{134}\text{Cs}$  in *I. galbana* and *P. tricornutum* on the basis of activity/cell

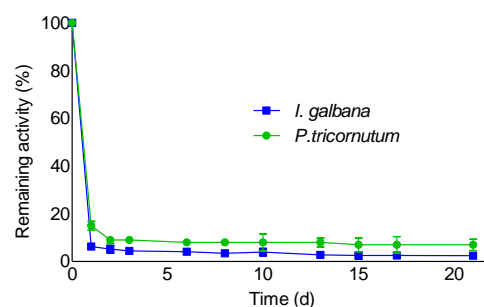


Fig. 3. The depuration kinetics of  $^{134}\text{Cs}$  after the dietary exposure (mean  $\pm$  SD; n=6).

The  $\text{DCF}_{ss}$  values were found to be  $5.8 \pm 2.0$  and  $6.2 \pm 2.0$  in *I. galbana* and *P. tricornutum*, respectively. Assimilation efficiency was found to be 6.3 and 15.1 in the clam following pulse chase feeding with *I. galbana* and *P. tricornutum*, respectively.

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## Establishing a base line for radioactivity in Alexandria, Egypt

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**Keywords:** natural radioactivity, marine environment, Solid State Nuclear Track Detector, Mediterranean coast of Egypt, beach sediments.

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Radioactivity in the marine environment is classified into natural and anthropogenic according to source. Most common long-lived radionuclides in the marine environment are U-238, Th-232 and K-40 and their decay series. Anthropogenic radioactivity includes artificial radionuclides such as Cs-137, strontium-90 etc. The presence of NORMs in the marine sediments depends on the mineralogy, grain size, biotic and some anthropogenic effects. Rn-222 attracts special attention among other nuclides due to its hazardous effect on human health.

Currently, The Egyptian Government is planning to build a nuclear power reactor at El Dabaa in the Mediterranean northwestern coast Frihy and El-Sayed (2012) which may comprise several impacts on the marine environment and public health. It is therefore crucial to establish a baseline for radioactivity levels (Ra-226, Th-232, K-40 and Rn-222) in the Mediterranean coast sediments and to link it to the sediment characteristics. This will enable assessing any change in the radioactivity level that might occur in the future. The study area passes through the three zones of the Egyptian Mediterranean coast representing three different sedimentary facies (figure1).



Figure 1: The study area, Alexandria, Egypt

Seventeen beach samples were collected from the study area. Sediment analyses included grain size analysis, heavy minerals separation and determination of total carbonate content. Radioactivity measurement was performed using sodium iodide scintillation counter to determine the concentrations of radionuclides (U-238, Ra-226, Th-232 and K-40). Radon activity measurement was determined using the solid state nuclear track detector (CR-39).

The results of radioactivity measurement are carried out using gamma spectroscopy Wang (2003), and Radon gas measurement using SSNTD Nikezic and Yu (2004), Fahmi *et al.*, (2008).

The results of radioactivity measurement show that the total radioactivity decreases westwards from Port Said to El Dabaa. Ra-226 activity concentration slightly increases westwards. The Ra-226 activity concentration is increasing with coarser mean grain size and total carbonates, while it decreases with increasing heavy minerals. The activity concentrations of Ra-226 in the Nile delta and Alexandria are attributed to the presence of heavy minerals. In the NW coast, uranium series is incorporated in the aragonite sediments during the formation of the chemically formed carbonates through physiochemical process. Considering the radiation dose hazard from the beach sediments, the average dose rate in the Egyptian Mediterranean coast is  $33.6 \text{ nGy h}^{-1}$ , which is less than the world average ( $55 \text{ nGy h}^{-1}$ ) suggested by the UNSCEAR. UNSCEA (2000).

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# Flooding evidence for the last 30 years in different types of lakes from Romania

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**Keywords:** *sediment dating, geochronology,  $^{210}\text{Pb}$  ( $^{210}\text{Po}$  method), flood comparison*  
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Several studies have been carried out regarding the determination of the geochronology and sedimentation rates in numerous types of lakes from Romania. However, none of these focus on the comparison of the sedimentation rates between the analysed lakes and the correlation of these with the major flood events.

This study has as an objective the comparison of both linear and mass sedimentation rates of four types of lakes (St. Ana Lake, Red Lake, Varsolt Lake and Matita Lake) situated in different geomorphologic areas of Romania, in order to determine the effects of the major floods on the catchment of the lakes.

The only existing volcanic lake in Romania is the St. Ana Lake. It is situated in the Eastern Carpathians, in the Ciomatu Mountains left to the Olt River. It is located on the bottom of an extinct volcano, Ciomatu, which last erupted more than four thousand years ago. Seven sediment cores were taken from this lake and it is to be noted, that the catchment area of the lake receives its water only by precipitation and runoffs along mountain slopes.

The Red Lake is a barrier lake in the Eastern Carpathians, formed in 1837 by the sliding and, respectively, collapsing of one of the slopes of the Ghilcos Mountain. This has obscured the valley of the Bicaz River in its confluence with the Suhard Creek. For retaining the high sedimentation, two dams have been built. A total of six sediment cores have been taken analyses from this lake (three from the lake itself, while other three from behind the dams).

Situated at the junction of the Apuseni Mountains and the Eastern Carpathians, the Varsolt Lake is the biggest lake in Salaj County. It is the source of fresh water supply for more than ten villages, as well as the seat of the county, Zalau. One sediment core has been analysed from this lake.

The Matita Lake is situated between the Chilia and Sulina branches in the fluvial part of the Danube Delta. While it receives water from several channels, the most water is supplied in the spring period, when the highest levels of the Danube are recorded. For this comparison, three sediment cores were used.

Each sediment core was sub-sampled into 1-3 cm layers. After weighting and drying them for 24 h at 75°C, physical parameters such as porosity, water content and bulk density were determined.

All sub-samples were analysed for  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  concentrations using high resolution gamma spectrometry, using an ORTEC GMX HPGe detector (FWHM of 1.92 keV at 1.33 MeV and a 0.5 mm Be window).

Alpha spectrometric measurements were carried out using an ORTEC SOLOIST 900mm<sup>2</sup> PIPS detector with a resolution of 19 keV coupled with an ASPEC-92 data acquisition system. The  $^{210}\text{Po}$  content of each sediment sample was determined using 0.5 g of dry sample and  $^{209}\text{Po}$  tracer for determination of chemical the yield. The dry sediments have been digested by a series of acids ( $\text{HNO}_3$ ,  $\text{HCl}$  and  $\text{H}_2\text{O}_2$ ), after which they have been deposited on high nickel content stainless steel discs (3 h at 85°C in a drying oven), interferents (iron ions) being eliminated by ascorbic acid. The geochronology of the sediments made with the  $^{210}\text{Pb}/^{210}\text{Po}$  method, using the CRS model.

After assessing each sediment core, several conclusions were taken. First of all, all analysed lakes have an increased sedimentation tendency. In case of the St. Ana Lake, the average quantity of past five of sedimentation has tripled, or as it happened in some parts of the lake, linear sedimentation has grown to values six times bigger (from 0.3 cm/y to 3.1 cm/y). Varsolt Lake is also showing an increasing tendency of sedimentation rates: from 1980 to 2002 mass sedimentation has shown a nearly linear yearly mass sedimentation growth of 0.025 g/cm<sup>2</sup>y.

Both Matita and Red Lake show more prominent changes in the sedimentation rates. In case of the Matita Lake the sedimentation rates show a constant growth until the early 2000's. Big increases in linear sedimentation rates can be observed in 2013 (14 cm/y and 4 cm/y) and in the period of 2009-2011 (9,5 cm/y and 6.5 cm/y), where two of Matita's sediment cores reach local minimums. Mass sedimentation rates are consistent with these, providing an average of 13,5 g/cm<sup>2</sup>y in 2013 and 13.15 g/cm<sup>2</sup>y for the 2009-2011 period. Six peaks can be observed in the 1980 ± 5 y period, having an average linear sedimentation rate of 6.5 cm/y. Similar data can be observed at the Red Lake: the year 2010 and the 2002-2005 period provide peaks in the linear sedimentation rates (4,75 cm/y and, respectively, 2.46 cm/y), while 2003-2005 and 1996-1998 periods provide increase in the mass sedimentation, causing average sedimentations of 2.67 g/cm<sup>2</sup>y and, respectively, 1.89 g/cm<sup>2</sup>y).

## Acknowledgments

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# **RESEARCH OF ARTIFICIAL RADIONUCLIDES DISTRIBUTION IN PLACES WHERE ULTRA LOW YIELD NUCLEAR EXPLOSIONS AND NON-NUCLEAR EXPLOSIVE EXPERIMENTS WERE CARRIED OUT**

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In the Soviet period Semipalatinsk Nuclear Test Site was a place of large scale works on testing nuclear weapons. Several testing spots are located at its territory that were used to perform different experiments with nuclear weapons in different environments: vertical boreholes, horizontal tunnels, in atmosphere and on ground surface. This work provides results of large-scale research performed at «Experimental Field » site, where the atmospheric and surface tests have been performed during the period from 1949 to 1963. Surface nuclear explosions were those lead to the most extensive contamination of the test site territory and the territory beyond it as well. In addition to explosions with nuclear energy extraction there is the information available on the fact that non-nuclear experiments, resulted in dispersion of significant amounts of nuclear materials in the environment were performed on the basis of «Experimental Field» infrastructure.

The main aim of this work is spectrometric survey of places where nuclear tests of super low yield and non-nuclear explosive experiments took place, reveal of epicenters and peculiarities of character and levels of contamination with artificial radionuclides in epicenters and fallout zones. Contaminated territories were researched using continuous and discrete pedestrian spectrometric survey, that allowed profiling with accuracy of 20 m and final detalization with resolution of up to 2.5 m respectively.

As a result of performed works it was determined that, super low yield nuclear explosions are characterized by a tight extended plumes with axial symmetry (length – up to 15km, width – up to 500 m), small crater or little to no crater is also a feature of such explosions. In addition, when these objects have been studied there was found no results of shock wave destructive effect and optical emission. In some cases, during inspection of objects of this type, chips and fragments of construction materials can be found in epicentral zone those get evaporated during explosions of higher yield. Contamination from ultra low yield explosions is characterized by high concentration of transuranium elements both in epicentral and fallout zone. At that, low concentration of fission and activation products or their virtual absence was found. When researching the P-2M site where 40 hydronuclear and hydrodynamical experiments was made, it was found that in general spatial distribution of contamination is of similar character, but its scale is significantly smaller, the length and the width of plumes do not exceed 5 and 0.2 km respectively. Products of contamination are dispersed transuranium elements and do not include fission and activation products.

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# Application of graphene oxide and magnetic graphene oxide for removal of radionuclides and heavy metals from contaminated wastewater

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**Keywords:** Pu, Am, heavy metals, sorption, magnetic graphene oxide.

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Graphene and related materials such as GO, reduced GO and various nanocomposites have attracted great attention since the graphene discovery in 2004. Their unique properties and a wide range of physical and engineering applications in various fields as well as their high potential for environmental remediation and for efficient removal of various pollutants including the most toxic long-lived radionuclides from contaminated solutions made them the most promising materials of the 21<sup>st</sup> century.

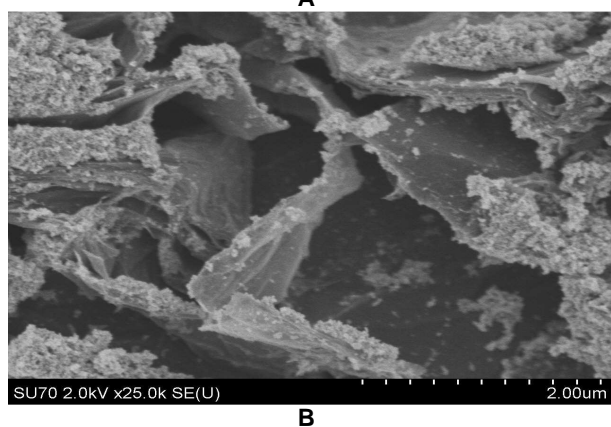
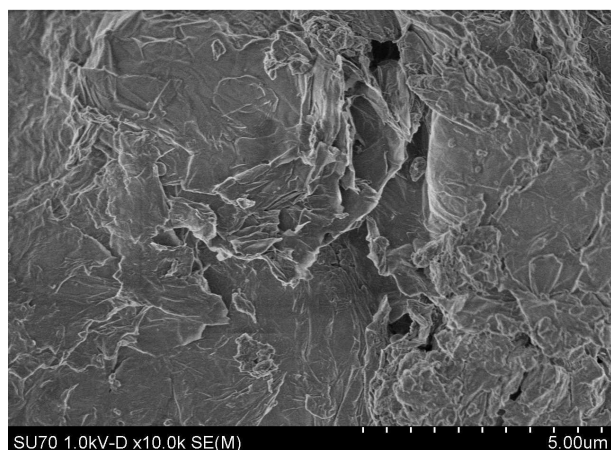


Figure 1. SEM images of GO (A) and MGO composite (B).

Graphene oxide (GO) and magnetic graphene oxide (MGO) composites with a different amount of magnetite were synthesized, characterized and used in sorption experiments.

Mössbauer spectroscopy, X-ray diffraction, Fourier transform infrared spectroscopy and Scanning electron microscopy (SEM) were used to characterize GO and MGO.

The effect of pH on sorption of Am(III) and Pu(IV) isotopes as well as Co(II), Ni(II), Cu(II) and Pb(II) to GO and MGO was studied in equilibrium and kinetic experiments. Large variations in the uptake of studied elements by adsorbents depending on the initial and final pH of solutions were observed.

The maximum uptake of Pu and Am by GO was found at the pH values of 3-5 and 7 for MGO with a low content of magnetite. Fast adsorption of all elements was observed and equilibrium was reached after 5 min of the contact time. About 100% of Ni was sorbed to GO at the initial pH of 3 to 8 after 24 h of contact time. Adsorption of Co gradually increased from 80% to 100% while the amount of adsorbed Cu and Pb decreased at pH of 7 and 8. For all elements the maximum adsorption was observed at initial pH values of 5 and 6. The obtained equilibrium adsorption data fitted well to the Freundlich isotherm model.

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# Radioactive characterization of Produced Waters from Gas Industry in Mexico

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**Keywords:** *Produced waters, NORM industry, Uranium, Radium*

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Mexico is a well-known gas producer, with important exploitations plants nowadays in operation. Historically, and in relation with this industrial activity, exists over the world some environmental concern due to possible chemical pollution. However, radiometrical characterization of some wastes and by-products of these gas extraction activities becomes also relevant as far as gas extraction is a well-known NORM (Naturally Occurring Radioactive Material) activity. Residues in the gas industry could have accumulations of natural radionuclides and hence, contribute to internal/external doses in humans, particularly during production, and maintenance, although some radiological environmental concern regarding disposal of generated waste should be taken also into account.

In association with the extraction of gas, huge amounts of the so-called “produced waters” are extracted and either disposed in artificial dams or used as a source of water for agriculture or other human activities. The chemical and radiometrical characterization of these waters should be performed periodically in order to evaluate their possible use.

In this work, produced waters from two different stations (Buena Suerte and Monclova stations) were collected and analyzed radiometrically and via ICP-MS. Both stations are located in the Mexican state of Coahuila, separated for less than 30 km and in each of them are collected the produced waters generated in more than 20 neighbouring production wells. The collected waters are then a mixture of the produced waters in several production wells.

These waters have been analysed chemically in a previous work (Martel-Vallés et al., 2014) resulting that in both cases the levels of total solids in dissolution are higher than the limits fixed in the Mexican legislation, while the values of the electrical conductivity are also quite high, being as a consequence only possible to use these produced waters in agriculture after its dilution. On the other hand, the water from Buena Suerte station presents high levels of hydrocarbons while the Monclova water overpass in Pb the toxic levels ( $\approx 5$ -1 mg/l).

In aliquots of these waters, the levels of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  have been determined by applying the gamma-ray spectrometric technique, while the levels of  $^{234}\text{U}$ ,  $^{238}\text{U}$  and  $^{210}\text{Po}$  have been determined by alpha-particle spectrometry after its sequential isolation through the application of a well-established

radiochemical procedure. Details of the radiometrical procedures can be found in (Mantero, 2013)

Radiometric results are compiled in Table 1, and deserve the following comments: Both waters can be considered NORM samples due to activity concentration levels clearly higher than the found ones in natural surface waters. In particular, levels of Ra-isotopes are quite high, while U-isotopes ones are lower. Activity concentrations in the samples are quite different, with clearly higher concentrations of Ra-isotopes and polonium in the Monclova sample (in association with the high levels of stable Pb that contains), while the concentrations of uranium are higher in the Buena Suerte sample (in association with its high levels of hydrocarbons). On the other hand, it should be highlighted the fact that in both samples exists a high disequilibrium between  $^{234}\text{U}$  and  $^{238}\text{U}$ , which is reflecting the origin of the waters and of the radionuclides in dissolution.

Table 1. Activity concentrations, in Bq/L, of several radionuclides from the U and Th series in the analysed produced waters. Between brackets are expressed the uncertainties in the determinations as one sigma.

Sample	$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{238}\text{U}$	$^{234}\text{U}$	$^{210}\text{Po}$
Buena Suerte	0.38 (0.05)	<0.30	0.047 (0.003)	0.074 (0.004)	0.017 (0.003)
Monclova	14.9 (0.3)	2.1 (0.1)	0.002 (0.001)	0.005 (0.001)	0.210 (0.007)

The obtained results indicates that that the radiological impact of the produced waters for the Mexican gas industry can be quite variable, in correlation with the variable levels of several radionuclides that can be found in the waters. An individual control of each station is needed for a proper management of these waters.

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## Tracer experiments in groundwater at new nuclear power plant site in Lithuania

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**Keywords:** Sodium fluorescein, salt, tracer, groundwater

Tracers tests in hydrology and water research defines the scientific field that aims at understanding the hydrologic system by making use of environmental and artificial tracers and modelling. Sodium fluorescein (uranine) and salt is the most popular tracers in the field due to its chemical properties, low detection limits and low costs.

Lithuania is planning to construct a new nuclear power plant (NPP) nearby the closed one. Groundwater characterization is an important issue in the setting process of new NPP. The tracer test has been started in 2013 year at new NPP site in Lithuania. The network was comprised of 19 wells, two systems. The first system defines in the first semi-confined intertill aquifer with depth of 10-19. The second system was installed in the unconfined aquifer (4.5-9 m depth). The sodium fluorescein and salt were chosen as a tracer. The concentration of sodium fluorescein was measured with the computer-controlled Aminco Bowman luminescence spectrometer and the salt was measured using ion chromatography system DIONEX ICS-5000. The results of test with tracers in groundwater show the flow and transport mechanism in this area, aquifer parameters (flow velocity and direction, effective porosity, hydraulic conductivity and etc.). The experiment results indicate the tracer's concentration in the wells water formed due to the water level fluctuations under natural conditions.

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# Temporal change of Pu inventory in water column of the Bering Sea

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Keywords: Pu isotope, inventory, water column, Bering Sea, temporal change.

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The anthropogenic radionuclides such as <sup>239</sup>Pu (half-life: 24,100 yr), <sup>240</sup>Pu (half-life: 6,560 yr) have been introduced to the surface oceans mainly as a consequence of atmospheric nuclear weapons testing. The dominant source of anthropogenic radionuclides in the early 1960s can be attributed to global stratospheric fallout from atmospheric nuclear weapons testing by the former Soviet Union. In the Pacific Ocean, there were significant contributions from close-in tropospheric fallout as a result of atmospheric tests on Bikini and Enewetak Atolls at the Pacific Proving Grounds (PPG) in the Marshall Islands (Yamada and Zheng, 2010).

Since the <sup>240</sup>Pu/<sup>239</sup>Pu atom ratio depends on the specific weapons design and test yield, it is a powerful fingerprint to identify the sources of Pu in the ocean (Buesseler, 1997). The mean <sup>240</sup>Pu/<sup>239</sup>Pu atom ratios from the global stratospheric fallout are  $0.176 \pm 0.014$  (Krey et al., 1976) or  $0.180 \pm 0.014$  (Kelley et al., 1999), whereas those from close-in tropospheric fallout from nuclear weapons testing at the PPG in the Marshall Islands are 0.33-0.36 (Buesseler, 1997).

The objectives of this study were to measure the vertical distributions of <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>239+240</sup>Pu concentrations and the <sup>240</sup>Pu/<sup>239</sup>Pu atom ratios in seawater from the Bering Sea, in order to trace temporal change of <sup>239+240</sup>Pu inventory in water column by comparing with measurements reported by the GEOSECS Expedition.

Seawater samples were collected at Stn. DR-13 in the Bering Sea with a double barrel PVC large-volume sampler. The <sup>240</sup>Pu/<sup>239</sup>Pu atom ratios were measured with a double-focusing SF-ICP-MS, which was equipped with a guard electrode to eliminate secondary discharge in the plasma and to enhance overall sensitivity (Zheng and Yamada, 2006).

Vertical profiles of <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>239+240</sup>Pu concentrations in the Bering Sea are shown in Fig. 1. The <sup>239</sup>Pu and <sup>240</sup>Pu concentrations were 9.9 and 8.6 mBq m<sup>-3</sup> in the surface water and decreased with depth; a broad maximum over the depth interval 200 – 700 m was observed, and the concentrations decreased with depth, then increased at the bottom layer. The subsurface <sup>239+240</sup>Pu maximum concentration was 16.3 mBq m<sup>-3</sup> at 400 m depth in the Bering Sea.

In order to elucidate the temporal variability in the vertical distribution of <sup>239+240</sup>Pu concentrations and inventories, distribution pattern of <sup>239+240</sup>Pu concentration

in the water column was compared between Stns. DR-13 and GX-219 (GEOSECS-219; 53°06'N, 177°17'W; sampling date: October 8, 1973) at the same latitude in the Bering Sea.

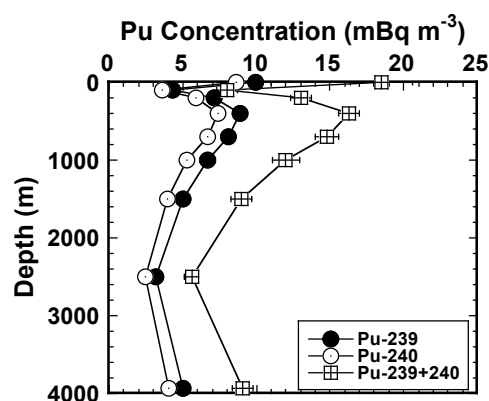


Figure 1. Depth profiles of <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>239+240</sup>Pu concentrations in the Bering Sea.

The <sup>239+240</sup>Pu inventories decreased from  $76.0 \pm 3.0$  Bq m<sup>-2</sup> at Stn. GX-219 to  $37.1 \pm 0.9$  Bq m<sup>-2</sup> at Stn. DR-13 during the period from 1973 to 1988. The difference in the <sup>239+240</sup>Pu inventories between Stns. DR-13 and GX-219 was  $38.9 \pm 3.0$  Bq m<sup>-2</sup>. The <sup>239+240</sup>Pu inventories decreased at average rates of  $2.6 \pm 0.2$  Bq m<sup>-2</sup> yr<sup>-1</sup> in the Bering Sea.

## Acknowledgments

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# Norm Assessment In Water Treatment Systems Poços De Caldas/BR Case

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NORM is the acronym used to refer to naturally occurring radioactive materials. Such materials can be used as raw material or turn into by-products, co-products or waste of several industrial activities. Oil and gas, mining and water treatment are examples of facilities that can handle NORM. In such cases, even if the radionuclides concentration in the raw material is low, concentration at significant levels from the perspective of environmental and occupational radiation protection may occur. Therefore, NORM presence and behavior in industrial production processes is a relevant subject of investigation and monitoring.

The Poços de Caldas Plateau presents what is called "radioactive anomalies", which are regions with natural radioactivity levels above those usually observed at the Earth's surface. A large hydrographic system permeates through the anomalous areas and also the region has high index of rain occurrence.

These anomalies, not surprisingly, host a uranium mineralization that, in the past, was explored as the first Brazilian uranium mining. Now in decommissioning process, the facility has waste rock piles where there is the occurrence of acid mine drainage (Oliveira, 1994).

Taking into account the characteristics of the region and the main aspects mentioned above, according to Palomo *et al* (2005), the local water treatment plants in operation can lead to production of radioactive waste as, for instance, sludge with accumulated radionuclides or contaminated filters.

This study aims to evaluate the presence of the species of the natural radioactive <sup>238</sup>U and <sup>232</sup>Th series in the treatment of city water elements Poços de Caldas - MG (water, materials and waste) and the possible need of radiation protection measures. Figure 1 summarizes the methodologies applied in the study.

Initial results showed that, although there is no significant presence of radionuclides in the treated water (supplied to the population), some radiological issues may arise from the wastes. One of the samples was the scale of the decanter tanks. The results obtained in the chemical and radiometric analyses.

Table 1. Scales (1<sup>st</sup> sampling) analyses results.

Sample	<sup>226</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>228</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>210</sup> Pb (Bq.kg <sup>-1</sup> )	U (Bq.kg <sup>-1</sup> )	Th (Bq.kg <sup>-1</sup> )
A	69,00 ± 17,0	71,00 ± 4,00	37 ± 2,00	< 505,63	< 202,20
B	164,00 ± 41,00	226,00 ± 11,00	30,00 ± 2,00	< 505,63	< 202,20
C	1.192,00 ± 298,00	1.704,00 ± 85,00	301,00 ± 15,00	< 505,63	< 202,20

The study can serve as an indication of the necessity of a more detailed investigation about this aspect and about the need of radioprotection measures in this kind of activity in the country. It is worth to mention that this is the first study of this kind carried out in the country.

## Acknowledgments

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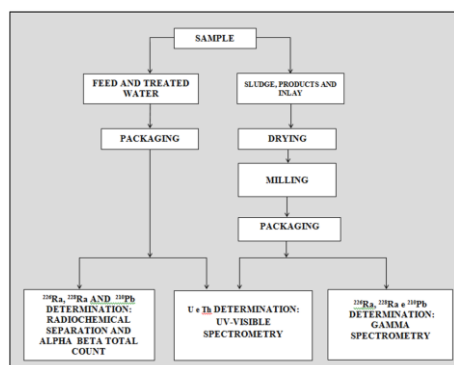


Figure 1. Sampling and analysis methodologies.

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## Three years monitoring of gamma-emitting radionuclides in fishing products sold in Italian market and arising from main FAO fishing areas

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After the Fukushima-Daiichi nuclear power plant accident in 2011, the requirement of checking the content of artificial radionuclides in fishing products has become relevant. Besides this more recent event largely advertised by media, many other potential sources of both natural and artificial radionuclides that can cause contamination of fishing products are present in the world: some of them are locally limited, others involve extended areas.

Among the sources of artificial radionuclides, there are the “acute” releases from accidents (such as Chernobyl) and from nuclear weapons tests in atmosphere, the low-level continuous controlled releases (usually locally limited) from nuclear power plants, research facilities and hospitals.

Among the sources of natural radionuclides, the “human enhanced” sources must be mentioned; they derive from many processes in non-nuclear industrial activities, such as mining, processing of phosphates, and coal power plants. These processes give rise to the production of substances, often wastes, with a so high concentration of natural radionuclides, such as <sup>226</sup>Ra, <sup>210</sup>Pb, and <sup>210</sup>Po, to become a risk from a radiological point of view.

In 2011, in the laboratories of IENI-CNR (Research Area of Padua), an extended research project for the monitoring of the content of gamma-emitting radionuclides in fishing products began. This project covered all the most traded fishing products in Northern Italy, and was performed through high resolution gamma spectrometry. The monitoring activity started just after the Fukushima-Daiichi NPP accident, and lasted 3 years, to individuate potentially contaminated fishing areas and possible phenomena of artificial radionuclides bioaccumulation. Samples of the more traded species (in Italian market) of fishes and molluscs were analysed; they became from all FAO fishing areas, were supplied

by a major wholesale dealer acting in Northern Italy, and are representative of all species bought from the same dealer and sold in the national market.

To reduce the counting time required by this analytical procedure, some “reference” radionuclides were chosen and used as “early warning” of contamination by artificial radionuclides. This allowed us to perform a screening on a very high number of samples (about 400). Moreover, activity concentrations of the most relevant natural radionuclides present in marine environment, such as <sup>40</sup>K, <sup>226</sup>Ra and decay products, were measured.

The values of the radionuclides concentrations so measured were used to individuate contaminated fishing areas and to evaluate the radiation doses for consumers.

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# Plutonium characteristics in sediments of Rømø coastal area in Denmark

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Keywords: Plutonium, <sup>137</sup>Cs, sediments, sources, Demark

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The long-lived anthropogenic radionuclides <sup>239</sup>Pu ( $t_{1/2}$ =24110 years) and <sup>240</sup>Pu ( $t_{1/2}$ =6561 years) have been released into the marine environment by human nuclear activities since the first nuclear weapons test in 1945. They are regarded as not only radioactive pollutants from a radiological viewpoint, but also useful tracers for better understanding of various physical and biogeochemical ocean processes due to their well-defined spatial and temporal inputs and unique chemical properties (Lindahl et al., 2010). With the source dependency of the Pu isotopic signature, the distribution and behaviour of Pu isotopes with different sources especially the Sellafield-derived Pu in the North Atlantic and its marginal seas have been intensively investigated in the past decades. However, observations on plutonium in sediments of Danish coastal area are relatively limited.

In this work, one sediment core collected in the coastal area of Rømø land in Denmark (55°8'815"N, 8°36'282"E) was analyzed for Pu isotopes using radiochemical separation combined with inductively coupled plasma mass spectrometry measurement to obtain information on the temporal variation of Pu activities, its isotopic composition and Pu inventory in the sediment column, in order to better assess the possible contamination of Pu and its characteristics in the area.

The analytical results of the sediment core showed that <sup>239+240</sup>Pu activities ranged from  $0.215 \pm 0.012$  to  $1.072 \pm 0.038$  mBq/g with an average of  $0.826$  mBq/g, while <sup>137</sup>Cs (decay corrected to 1<sup>st</sup> Sept. 2012) ranged from  $2.40 \pm 0.39$  to  $49.52 \pm 1.39$  mBq/g with an average of  $31.08$  mBq/g. It can be observed from the Figure 1 that the concentrations of <sup>239+240</sup>Pu and <sup>137</sup>Cs show very similar increasing trend from the surface sediment to 50 cm depth. No bottom background levels of both <sup>239+240</sup>Pu and <sup>137</sup>Cs activities were observed in the sediment core. The most possible explanation is that the sedimentation rates in the sampling site is very high so that the sampling depth of 50 cm is not long enough to detect the background level, and the layer below 50 cm depth in the sampling site should contained some part of radionuclides depositions. The inventory of <sup>239+240</sup>Pu with the limited 50 cm depth in the core was calculated to be  $172$  Bq/m<sup>2</sup>, which was much higher than the global fallout value of  $\sim 50$  Bq/m<sup>2</sup> for 50~60N, suggesting that significant amount of Pu injection into the sampling region in the past decades.

The <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios in the sediment core (Fig. 2) ranged from 0.179 to 0.198 with an average of  $0.189 \pm 0.005$ , relatively constant and close to the global fallout value ( $0.180 \pm 0.014$ , Kelley et al., 1999) and the

value of Sellafield derived Pu (Cooper et al., 2000) for recent years. While <sup>241</sup>Pu/<sup>239</sup>Pu atomic ratios (corrected to 16<sup>th</sup> May 2013) were also relatively constant in the core with an average of  $0.00335 \pm 0.00174$ , generally higher than the global fallout value of 0.0011 but comparable with the Sellafield-derived Pu ratio of 0.0048, indicating that the remobilization and redistribution of Pu from Sellafield discharges should contribute in some part to Pu contamination in the coastal area of Denmark in addition to river-in global fallout.

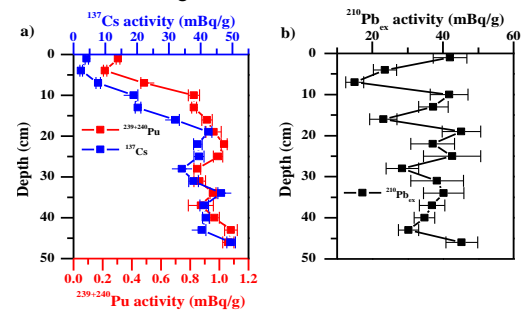


Figure 1. Vertical distribution of <sup>239+240</sup>Pu, <sup>137</sup>Cs and <sup>210</sup>Pb<sub>ex</sub> activities in the sediment core 2012-1333.

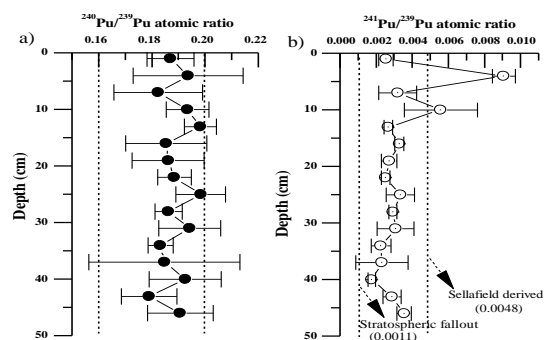


Figure 2. Vertical distribution of atomic ratios of Pu isotopes in the sediment core 2012-1333.

## Acknowledgments

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# Method for Organically Bound Tritium analysis from sediment using a combustion bomb

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**Keywords:** organically bound tritium, sediments, liquid scintillation counting, Parr bomb.  
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Current measurement of organically bound tritium (OBT) from sediment using liquid scintillation counting (LSC) implies first of all conversion of organic matter in liquid water. In order to convert sediment organic matter to water it is a necessity to perform a total oxidation process, which most commonly can be made through combustion using different type of tube catalytic furnaces (Cossonnet, 2009; Eyrolle-Boyer, 2014) or the devices called oxidizers (Kim, 2013).

The objective of this study was to develop, optimise and verify a new method for analysis of OBT from sediments, using a Parr bomb type 1121, a versatile device needing limited financial resources, already available in our laboratory. The content of carbon and hydrogen in sediments are quite low, around 3% for carbon and 1% or less for sediments, and the combustion using a Parr bomb has limitations. In order to use this kind of device for sediment combustion we needed to mix the sample with a tritium free combustion promoter, in our case a dried heavy fuel oil. The preliminary experiments involved the use of two fossil tritium free combustion promoters, a heavy fuel oil (petroleum derivate) and a coal. Between the two promoters the heavy fuel oil was selected for its high viscosity, which ensures good homogeneity of the mixture and prevents the deposition of the sediment on the bottom of the combustion bomb capsule.

The experimental setup implied mixing in different ratios of grounded dried sediment with the heavy fuel oil, directly into the combustion capsule of the combustion bomb. The ratio between sample and fuel oil varied from 1/3 to 1. The obtained combustion water was collected directly from the combustion bomb, neutralized and then purified using chemical treatment ( $\text{Na}_2\text{O}_2$  and  $\text{KMnO}_4$ ) followed by lyophilisation before measurement using LSC. All tritium measurements were performed with a Quantulus 1220 using 8 g of purified combustion water and 12 g of uLLT scintillation cocktail from Perkin Elmer. In order to obtain sufficient amount of water we combusted mixtures of 12 g heavy oil (with a hydrogen content of 9.55%) and 4, 6, 8, 10 and 12 g of sediment (with a hydrogen content of 0.79%).

Due to the use of heavy fuel oil, tritium from the sediment was diluted during the combustion, dilution factors depending on the sediment: heavy fuel oil ratio. In Table 1 we presented the dilution factors for each of the performed experiments and also OBT activity measured in combustion water of the sediment, expressed in Bq/L.

Table 1. Dilution factors  $f_{dil}$  for different ratios of sediment: heavy oil used in combustion bomb and OBT activity measured in the sediment.

Sediment : Heavy fuel oil [g:g]	Dilution factor $f_{dil}$	Tritium activity in sediment [Bq/L]
4:12	37.2658	$30.13 \pm 7.82$
6:12	25.1772	$30.90 \pm 5.45$
8:12	19.1329	$26.56 \pm 4.19$
10:12	15.5068	$24.68 \pm 3.45$
12:12	13.0886	$17.28 \pm 2.86$

The obtained results from OBT analysis in the studied sample were not the same, although a single sample of sediment was analyzed. This fact is due to incomplete combustion of organic matter in the sample for higher ratios sample: heavy oil, having as result a dilution of OBT activity.

In conclusion the new developed method can be used to OBT measurement in sediment. The proper ratio sample: heavy oil in our experiments was between 1/3 and 1/2, higher ratio leading to unreliable results, due to incomplete combustion of the sample. The limitation of this method is that the low levels of OBT in sediment can not be measured, due to the tritium dilution during combustion, minimum measurable activity being around 10 Bq/L.

This method was used to measure the estuarine sediment sample from the 2<sup>nd</sup> OBT Intercomparison Exercise and the sample: heavy oil ratio used was 6:12 (wt.), the reported value being  $163.16 \pm 11.55$  Bq/kg.

## Acknowledgments

This work was performed within the framework of the Experimental Pilot Plant for Tritium and Deuterium Separation from Institute for Cryogenics and Isotopic Technologies.

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## Risk-informed prioritization and systems-based approaches for remediation of deep subsurface contamination

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Systems-based approaches are used within a structured framework to describe contaminant behavior along potential exposure pathways in a way that facilitates an understanding of interfaces—such as those between the vadose zone and groundwater, within aquifer systems, and at discharge locations. Within this framework, controlling subsurface features and processes are identified and related to potential exposure pathways. This understanding enables (1) evaluation and quantification of the risks posed by subsurface contamination to human health and the environment and (2) more effective design of cost-effective, systems-based remediation and monitoring strategies focused on establishing and maintaining protectiveness and mitigating future risk through appropriate remedial actions.

In situ remediation approaches are being evaluated at complex sites as potential options to address contaminants in the deep subsurface, including the vadose zone and groundwater. These techniques mitigate risk and protect human health and the environment, but may leave contaminants in place.

Some of the candidate approaches for the vadose zone are based on changing the contaminants or subsurface conditions in a way that slows downward migration of the contaminants using amendments delivered in the gas-phase. Two promising approaches

that have been tested are soil desiccation and ammonia-induced sequestration of contaminants.

For soil desiccation, a dry gas is injected to desiccate a targeted portion of the subsurface and thereby decrease contaminant movement by removing moisture and decreasing the hydraulic conductivity of the desiccated zone. Ammonia-induced sequestration of contaminants relies on changing the pore water chemistry, primarily through pH changes, to induce dissolution and precipitation processes that decrease the amount of mobile contaminants in the vadose zone.

In the groundwater, in situ remedies have become relatively common. However, due to cost and access restrictions in the deep subsurface, advances in remediation are emphasizing more targeted approaches that enhance natural processes to accomplish overall remediation goals. These remediation approaches require a strong understanding of the subsurface system and natural attenuation factors to enable a focus on controlling features or processes in designing the remediation approach. Integration with monitoring approaches and designing with the capability to adapt remedies are important for deep and complex contaminant issues where plume behavior and responses to actions are difficult to predict at the onset of site characterization and remedy selection activities. This holistic approach enables and supports long-term, scientifically defensible remediation decisions



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# AMAD of $^7\text{Be}$ aerosol under different meteorological conditions and different environments

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The activity size distributions of the natural radionuclide tracer  $^7\text{Be}$  in different size fractions (<0.4  $\mu\text{m}$ , 0.4-0.7  $\mu\text{m}$ , 0.7-1.1  $\mu\text{m}$ , 1.1-2.1  $\mu\text{m}$ , 2.1-3.1  $\mu\text{m}$ , 3.1-4.2  $\mu\text{m}$ , 4.2-5.8  $\mu\text{m}$ , 5.8-9.0  $\mu\text{m}$ , >9.0  $\mu\text{m}$ ) were determined at different site places in Northern Italy. Samplings were carried out during the four different seasons of the year 2011. The aim of this work was to define any differences due to the different environments and different meteorological conditions and clarify the main parameters influencing the activity size distribution of radioactive aerosols.

Four different locations were chosen for sampling, (a) a suburban - industrialised area (Segrate, Milan) – the reference station, (b) an urban area in downtown Milan (c) a rural-residential area (Ispra) and (d) a rural area at Monte Rosa mountain (1300 m asl). The first station at Segrate, Milan has been chosen as a reference station, so each sampling at the other three sampling stations was carried out simultaneously with the sampling at the reference station.

The total experimental collection period lasted almost a full year (from February 2011 to December 2011), covering all seasons of the year 2011. The length of each collection period was 7-10 days with regulated air flow rate of about 28.3 L min<sup>-1</sup> (1 cfm) and collected air volume of about 300-400 m<sup>3</sup>. All samplings have been carried out by two compatible 1ACFM 9-stages cascade impactors and with Efficient Cutoff Diameters (ECD) of 0.4, 0.7, 1.1, 2.1, 3.3, 4.7, 5.8, and 9  $\mu\text{m}$ . Acetate Cellulose filters were used as collection substrates (0.8 mm pore size Sartorius – Germany of 8.2 cm diameter). No additional coating (oil or grease) was used.

At the end of the collection procedure, the filters used as plane sources were measured for  $^7\text{Be}$  activity (E<sub>g</sub> = 477 keV) using two high resolution, high relative efficiency (42%), low-background HPGe detectors. The statistical uncertainty ranges from about 12%-18% for the activities on the impactor stages 5-9 (finer particles) to about 20-30% for the activities on the impactor stages 1-4 (coarser particles).

Analysis of data gave that the greatest part of  $^7\text{Be}$  aerosols is associated with fine particulate.

The activity median aerodynamic diameter (AMAD) ranged from 0.40  $\mu\text{m}$  to 1.05  $\mu\text{m}$  during winter period, between 0.50-0.73  $\mu\text{m}$  during spring period, between 0.47-0.69  $\mu\text{m}$  during summer period and between 0.50-0.96  $\mu\text{m}$  during autumn period.

Lower AMAD values are recorded during summer period which in general is characterized by low relative humidity conditions and high temperatures. During winter period the highest AMAD values are recorded. These results support the results of previous investigations reported in the literature for entirely different environments and under different meteorological conditions (Ioannidou, 2011, Papastefanou and Ioannidou 1996, Yu and Lee 2002).

At all stations, except one in Ispra region near the lake Magore, the AMAD values were anticorrelated with  $^7\text{Be}$  activities, while they are correlated with RH%. An anti-correlation was observed between the AMAD values and  $^7\text{Be}$  activities. The observed correlation between the AMAD values and RH% can be due to the intense condensation process during high RH% conditions resulting in increased particle sizes and higher scavenging rates of aerosols.

Finally, in Segrate region and in downtown stations, which are located in high polluted environments, the AMAD values of  $^7\text{Be}$  aerosol particles are greater than AMAD values in less polluted environments and far from industrial activities regions of ISPRa and Monte Rosa Mountain. These results, combined with the reported PM<sub>2.5</sub> and PM<sub>10</sub> in the regions of investigation, support our assumption that under polluted environments the radioactive aerosol particles are connected with greater aerosol particles. So the radioactive nuclides can serve as an index of air polluted conditions.

To the best of our knowledge this is the first study of activity size distribution of a radioactive nuclide simultaneously at different environments and during the four seasons of a year.

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# Simultaneous determination of specific alpha and beta emitters by LSC-PLS in water samples

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Keywords: *alpha emitters, beta emitters, liquid scintillation, water samples*  
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The extended abstract should be in double column (like this example) and ONE page long. Use A4 page set-up and make all margins (top, bottom, left, right) 2.5 cm wide. Use 9 pt Helvetica fonts (except for the title which should be in 12 pt bold and in sentence case).

Liquid scintillation counting (LSC) is meaningful technique for the determination of alpha and beta emitters. Otherwise poor resolution, especially for beta emitters hinders the simultaneous determination of several alpha and beta emitters from the same spectrum. However, to achieve simultaneous determination of several beta emitters, by just one assay consisting of an easy and fast sample treatment and subsequent measurement by liquid scintillation counting, makes it possible to avoid expensive and time consuming radiochemical separations. Because of this, several authors studied and proposed different approaches based on multiple windows definition, spectra deconvolution or chemometric techniques, to identify and quantify single isotopes in complex radionuclide mixtures.

In this work liquid scintillation spectrometry with Partial Least Square (PLS) calibration was used to determine different alpha ( $^{nat}U$  and  $^{241}Am$ ) and beta emitters ( $^{40}K$ ,  $^{60}Co$ ,  $^{90}Sr$ / $^{90}Y$ ,  $^{134}Cs$  and  $^{137}Cs$ ) in water samples. The isotopes selected cover a wide energy range of both, alpha emissions (from 4.20 MeV of  $^{238}U$  to 5.49 MeV of  $^{241}Am$ ) and beta emissions (from 0.31 MeV of  $^{60}Co$  to 2.28 MeV of  $^{90}Y$ ).

The analytical procedure used is based on evaporate to dryness a 100 mL aliquot of the sample. The precipitated obtained is dissolved in 10 mL of deionised water acidified by HCl to pH=1.5. Afterwards 8 ml aliquot of this dissolution is mixed with 12 ml of Ultima Gold AB in PE vials. The vial is counted with the ultra-low level liquid scintillation spectrometer QUANTULUS

1220, after 2 hours remaining in darkness to avoid photoluminescence phenomena.

Two PLS models were tested. One (Model A) calibrated with 6 radionuclides ( $^{nat}U$ ,  $^{241}Am$ ,  $^{40}K$ ,  $^{60}Co$ ,  $^{90}Sr$ / $^{90}Y$ , and  $^{137}Cs$ ), and the other (Model B) with 7 radionuclides including  $^{134}Cs$ . The models were tested with spiked mixtures of isotopes. The error of prediction of both models was evaluated around 10 % for almost all the radionuclides. For  $^{137}Cs$  this error was around 30 %. In cases were the sample contains an interferent not included in the calibration set ( $^{134}Cs$  for Model A) the error of prediction for  $^{137}Cs$  was unacceptable (115%).

## Acknowledgments

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# Detection of Ra-223 from therapeutic applications in municipal sewage sludge

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The alpha-emitting radioisotope Radium-223 (Ra-223) has recently been established for the treatment of metastatic prostate cancer (Pandit-Tasker et al., 2014). Ra-223 is an alpha-emitter with a half-life of 11.4 days. In 2013 a clinical study was conducted with a restricted number of patients in several Swiss hospitals. The patients were administered a total activity of 4.2 MBq Ra-223 in an ambulatory treatment, after which the patients returned to their homes.

With the patient's excretions a part of the Ra-223 will find its way into the sanitary sewer and further downstream into the waste water treatment plant. Radium has a strong affinity towards adsorption on iron-(hydr)oxides and organic matter. Since iron-hydroxide precipitation is an important step in the wastewater treatment in all the studied plants it seems likely that Ra-223 finally becomes captured into the sewage sludge.

In order to verify this first guess and in order to determine the actual concentration levels of Ra-223 from medical applications in sewage sludge we analysed sewage sludge from waste water treatment plants with Ra-223-treated patients living in the drainage area.

HPGe- $\gamma$ -spectrometry measurements of 500 g of sample for 80'000 s resulted in typical detection limits of 0.5 Bq/kg or less.

Preliminary mass balance calculations suggest that Ra-223 is indeed largely transferred to the sewage sludge. Concentrations of up to several Bq/kg were observed in sewage sludge of a plant with one treated patient living in its drainage area. Therefore, even with several patients living in the same area the Swiss exemption level of 100 Bq/kg for Ra-223 is unlikely to be exceeded.

## Acknowledgments

For fruitful discussions as well as help with sampling and analytical work and the author thanks Gloria Perewusnyk and Giovanni Ferreri.

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# Correlations between soil characteristics and radioactivity content of Vojvodina soil

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The intensity of the processes of soil sorption/desorption, migration, retention and translocation is influenced by the nature of the given radionuclide, the type of soil and of crops grown on it, and the climatic conditions.

During the year 2001, the radioactivity of the soil in Northern Province of Serbia – Vojvodina was measured (I. Bikit, et al. 2005). The main conclusion was that there has not been any increase of radioactivity that could endanger the food production present. During the year 2010, Laboratory for radioactivity and dose measurement in cooperation with the Institute of Field and Vegetable Crops repeated radioactivity monitoring of agricultural soil on the same 50 locations (Figure 1). In this paper we compared the obtained results from 2001 and 2010.



Figure 1. The map of Vojvodina with 50 locations of sampling

The locations of these 50 samples were selected so that they proportionately represented all geomorphological units (Koščál et al., 2005): two mountains, four loess plateaus, three loess terraces, four alluvial plains, two sandstone terrains and all soil types (IUSS Working Group WRB, 2014): Chernozem, Vertisol, Fluvisol, Cambisol, Planosol, Solonchak and Solonetz. The content of clay and humus varied within wide limits depending on soil type.

A simple method developed in Novi Sad laboratory was used for the determination of the <sup>238</sup>U activity concentration from gamma-lines of the first progeny of this radionuclide, <sup>234</sup>Th. The samples of soil were measured on GMX type HPGe germanium detector with extended range (10 keV - 3 MeV) in lead shield of 12 cm wall thickness. The gamma spectra were acquired

and analyzed using the Genie 2000 software. All measurement uncertainties are presented at 95% confidence level.

Table 1. Radionuclide content ranges for different years and depths.

radionuclide	A [Bq/kg]		
	2001 0-5 cm	2010 0-5 cm	2010 30 cm
<sup>238</sup> U	24 - 72	9,4-80	11,5 - 87
<sup>226</sup> Ra	19,1 - 51	9,7-49,1	12,4 - 44,7
<sup>232</sup> Th	22 - 62	11,7-70,5	22 - 55,5
<sup>40</sup> K	238 - 730	238-1000	312 - 783
<sup>137</sup> Cs	5,7 - 55	3,0-42,6	3,1 - 29

This study confirmed that the content of detected radionuclides in soil which was sampled from the surface layer, differed only within the measurement error from the content of radionuclides in soil sampled from a depth of 30 cm, i.e. depth within rootsystems of most agricultural crops is placed. This conclusion can be applied in developing and establishing the method of soil sampling for radioactivity determination.

The possibility of soil contamination by use of fertilizers with a high content of uranium <sup>238</sup>U and the safety of food production are also analyzed in this paper. In all soil samples potassium <sup>40</sup>K bounded to clay-like structure of Vojvodina soil was detected depending on crops and organic residues that remain in the soil after the harvest.

## Acknowledgments

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# Influence of soil texture and availability on the soil-to-plant transfer of $^{238}\text{U}$ and $^{226}\text{Ra}$

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**Keywords:** natural radionuclides, soil-to-plant transfer, soil texture, availability.

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The soil-to-plant transfer factor was determined for the long-lived radionuclides of the uranium series,  $^{238}\text{U}$  (and  $^{234}\text{U}$ ) and  $^{226}\text{Ra}$ , in a granitic area in the south-west of Spain. This area presents elevated concentrations of these radionuclides due to its location next to a disused uranium mine (Vera Tomé *et al* 2002). The plant compartment comprised the aboveground fraction of herbaceous species characteristic of a Mediterranean area. For the soil compartment, different options were considered. A first study was carried out considering the bulk soil and its labile (available) fraction as soil compartments. Then, the bulk soil was divided into three granulometric fractions: coarse sand (0.5–2 mm), fine sand (0.067–0.5 mm), and silt and clay (<0.067 mm), and the soil-to-plant transfer was again studied considering these three texture components as soil compartments. Finally, the labile fraction was extracted from each texture component, and the activity concentrations of the radionuclides studied were determined in each fraction.

The activity concentrations of the uranium and radium isotopes in the different soil fractions and the vegetation samples were determined by alpha-spectrometry with 450 mm<sup>2</sup> active area PIPS semiconductor detectors.

In order to assess the influence of soil texture and availability on the soil-to-plant transfer process, possible correlations were sought between the activity concentrations of each of the soil compartments and that of the plant compartment, using in all cases the log-transformed data. Thus, a linear correlation with a slope indistinguishable from unity would be indicative of proportionality between the two sets of untransformed data.

The results showed the uranium concentrations in the herbaceous plants to be strongly log-log correlated with those in the bulk soil ( $r=0.998$ ;  $p=0.044$ ). When the different granulometric fractions were considered as substrate, the relation between the herbaceous and the substrate uranium concentrations presented different behaviours. In the case of the coarse sand fraction, the log-transformed concentrations showed no statistically significant correlations. But for the fine sand fraction, a correlation coefficient of  $r=0.994$  was obtained ( $p=0.07$ ), with a slope of the linear regression of  $1.25\pm0.14$ . The result was even stronger with the finest fraction (limes and clays), for which the correlation coefficient was

$r=0.995$  ( $p=0.06$ ) and the slope was statistically indistinguishable from unity ( $1.0\pm0.1$ ). Both of these slopes are consistent with proportionality between the plant compartment activity concentration and that of the respective soil fractions. When the available fractions were considered, strong correlations were found between the herbaceous concentration and the available concentration of the three texture components considered. The correlation was especially strong for the coarse-sand available fraction ( $r=0.999$ ;  $p=0.006$ ).

For the radium concentrations, the log-log correlation of the plant compartment with the bulk soil ( $r=0.823$ ) was not statistically significant at a 90% confidence level. The correlation was better when only the available fraction was considered as substrate ( $r=0.934$ ,  $p=0.066$ ), with a slope of  $0.84\pm0.23$ . This finding implies that the soil-to-plant transfer factor ( $TF$ ) can be considered constant and independent of the substrate concentration when the bulk-soil available fraction is considered as substrate. For the granulometric components, there were linear log-log relationships between the radium concentrations in the plant compartment and those in the different fractions were linear in all cases, but with statistical significance at a 90% confidence level only with the coarse sand fraction and the limes-and-clays available fraction as substrates. The slopes in both these cases were statistically indistinguishable from unity, indicative of proportionality between the (untransformed) radium concentrations in the plant component and the respective substrates. In these cases, the respective substrate could also be used to redefine  $TF$ . The geometric means in these two cases were  $1.51\pm0.17$  and  $16.6\pm1.9$  for the coarse sand fraction and the limes-and-clays available fraction, respectively, with the uncertainties in both cases being just greater than 10%.

## Acknowledgments

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# INVESTIGATION OF DISTRIBUTION OF RADIOACTIVE CONTAMINATION IN THE "WATER – SEDIMENTS" SYSTEM OF THE SEMIPALATINSK TEST SITE AND ADJACENT TERRITORIES

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For complex assessment of condition of surface water objects (water reservoirs and streamflows) it is very important to have an information about radioactive contamination of both water and sediments. Accumulating contamination during a long time, sediments can serve as an indicator of radioecological state of the water object. Herewith, they can accumulate radionuclides and be the source of secondary radioactive contamination of the water object [1]. The aim of this work is to study the character of distribution of radioactive contamination in the "water – sediments" system of objects of Semipalatinsk Test Site (STS) and adjacent territories.

As the research objects there were chosen water reservoirs and streamflows of "Experimental Field", "Balapan", "Degelen", "Telkem" sites, adjacent territories as well as Shagan river. At selected objects conjugated samples of water and sediments were taken. Samples were taken at the distance of 1 – 2m from the costal line, and as mainly all the water objects are not deep-water ones, the depth in sampling points was about 20 – 30 cm. Sampling of sediments was made to the depth of 0 – 10 cm, water – from the surface 0 – 20 cm. Collected samples were used to determine concentration of artificial radionuclides  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ .

As a result of performed researches transfer factors (TF), determining character of distribution of radioactive contamination in water objects were calculated.

Table 1. TF for water objects of STS

Sampling location	TF			
	$^{241}\text{Am}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{239+240}\text{Pu}$
"Experimental Field" site	$> 2 \cdot 10^4$	$> 3 \cdot 10^5$	$> 1 \cdot 10^3$	$4 \cdot 10^6$
«Atomic» lake	$> 2 \cdot 10^2$	$> 2 \cdot 10^5$		
Water reservoir	$> 8$	$> 3 \cdot 10^3$		
"Telkem" site		$> 7 \cdot 10^4$	$> 1 \cdot 10^3$	
Shagan river		$> 4 \cdot 10^3$		
"Degelen" site		$> 1 \cdot 10^3$		
Adjacent territories		$> 2 \cdot 10^5$	$> 3 \cdot 10^3$	$> 2 \cdot 10^6$

According to obtained data the TF is decreasing in the range  $^{239+240}\text{Pu} > ^{137}\text{Cs} > ^{241}\text{Am} > ^{90}\text{Sr}$  i.e.  $n \times 10^6 > n \times 10^5 > n \times 10^4 > n \times 10^3$  respectively. However, obtained TF is  $\gg 1$ . It indicates that, the major part of investigated radionuclides in the "water – sediments" system is concentrated in sediments.

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# Fallout measurements In North Italian Regions after Fukushima Nuclear Power Plant Accident

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**Keywords:** Fukushima accident; radionuclides fallout; Cs-137; milk.

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Radionuclides released during an accident can be transported over long distances, before they are deposited by fallout on vegetation and soil. Such an event occurred after the Chernobyl accident in April 1986 where extensive fallout of various long- and short lived radionuclides was observable in many European countries, including Italy. The resulting contamination of the vegetation has decreased due to radioactive decay and weathering. Longer lived radionuclides, however, are still present in the soil and are taken up by plants via their roots. In particular <sup>137</sup>Cs that is one of the most important contaminants because of its long half life, affinity for biological system and its uptake to man through diet. In March 2011 Fukushima accident contributed with new input in the atmosphere of contaminants: a cloud containing radioactivity was formed in air over Fukushima nuclear power plant NPP, moved over the Pacific Ocean in the direction of the Arctic Ocean, enter in the Atlantic Ocean and diffused over the European continent. The transferred radioactivity was detectable all over the world: many laboratories started measurements and presented results on the fallout from the Fukushima nuclear accident<sup>1</sup>. Also in Italy were made a series of measurements of the fission product radionuclides in different regions and places. In particular our research group was focused on measurements of radioactivity present in air, water, snow, soil, grass and milk in Milano and Monte Rosa region.

The results of these measurements are justified by the carried out back trajectories analysis, that describes the path of the air masses transportation from Fukushima since Milano area.

In fresh goat and cow milk produced and collected in a farm at a village in Anzasca valley near Macugnaga (rural area), in Monte Rosa mountain at 500 m height in the period April to July 2011 were found small values of concentration of <sup>131</sup>I and <sup>137</sup>Cs. The <sup>134</sup>Cs/<sup>137</sup>Cs activity ratios can reveal information on radioisotope origin. The relative high activity concentrations of <sup>137</sup>Cs and the very low values of the activity ratio of <sup>134</sup>Cs/<sup>137</sup>Cs, in combination with the absence of <sup>134</sup>Cs in most of the cases indicate a strong contribution from "older" <sup>137</sup>Cs, probably from the Chernobyl accident and past global fallout and not from Fukushima accident. After these preliminary results we start on January 2013 a campaign in order to continue to monitor this area and have some reference data of radiocaesium concentration level due to different fallouts. Preliminary results of two time series data collected are presented in Figure 1.

Some measurements were conducted in order to find a correlation between the <sup>137</sup>Cs concentration values and the animals pasture and environments which are utilized for food production (due to the pathway: fodder-animal-milk).

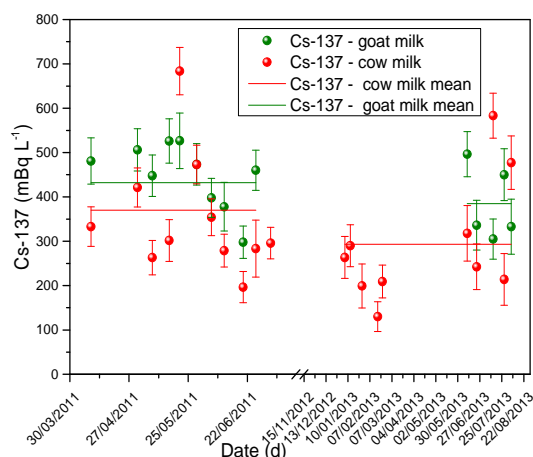


Figure 1. <sup>137</sup>Cs concentration in cow and goat milk collected in a farm in Monte Rosa region.

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# RADIOACTIVITY LEVELS OF MAIZE AND DOSE ESTIMATES FOR THE PUBLIC IN TANZANIA

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## **Abstract**

Natural radioactivity levels in maize which is one of the staple foods in various regions in Tanzania have been studied. The radioactivity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were determined using  $\gamma$  ray spectrometry employing HPGe detector of relative efficiency of 51 %. The average radioactivity concentrations in maize from four regions were ranged from  $3.2 \pm 0.2$  to  $25.6 \pm 0.7$  Bq/kg  $^{238}\text{U}$ ,  $4.9 \pm 0.3$  to  $72.9 \pm 1.0$  Bq/kg for  $^{232}\text{Th}$  and  $32.5 \pm 7.2$  to  $434.6 \pm 18.7$  Bq/kg for  $^{40}\text{K}$  respectively. Total annual committed effective dose due to total  $^{238}\text{U}$  and  $^{232}\text{Th}$  intakes as a result of consumption of maize in four Regions were as follows; Manyara (1.05 mSv/y), Mbeya (0.12 mSv/y), Ruvuma (0.07 mSv/y) and Dar es Salaam (0.06 mSv/y). The dose values are lower than the annual dose guideline for the general public which is 1 mSv/y, except the dose value from Manyara.

**Key words:** Radioactivity, Minjingu phosphate fertilizer, committed effective dose.

# DISTRUBIOTION OF ENVIRONMENTAL RADIONUCLIDES IN THE BUILDING MATERIALS AND RELEATED HEALTH RISK ASSESSMENT

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**Keywords:** Building material, NORM, Indoor Radiation, Health Risk assessment, HPGe

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Humans are being exposed to the background radiation from natural environment every day; however, the main contributor of human exposure to natural radiation is the indoor.

Due to the radiotoxicity of terrestrial natural radionuclides in the soils, a raw material in the building industry, the activity of main naturally occurring radionuclides (<sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K) in 23 common building materials, use in Mahallat, Iran, were determined to estimate the hazard index and assess the health risk of indoor gamma radiation.

The High-purity Germanium detector (HPGe), with low background, high resolution and high efficiency, was used to detect gamma ray emission from radionuclides present in the samples. The absolute efficiency of each energy peak was calculated using a mixed radionuclides gamma standard soil (IAEA-326).

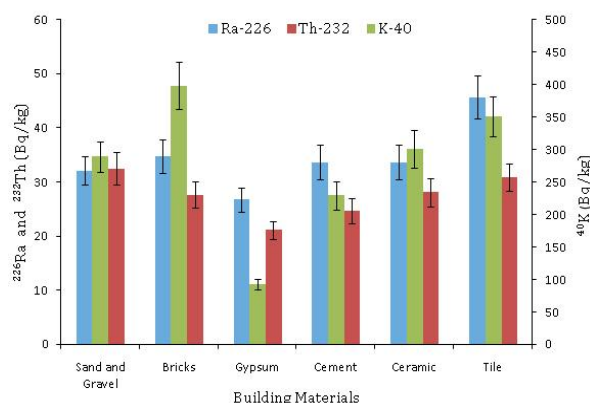
The activity concentrations of <sup>232</sup>Th, <sup>226</sup>Ra and <sup>40</sup>K were measured in the range of 18±1 to 44±4 Bq/kg (27±2 Bq/kg), 22±2 to 53±4 Bq/kg (34±3 Bq/kg) and 82±8 to 428±37 Bq/kg (275±24 Bq/kg), respectively.

The average radium equivalent activity and the external hazard index were calculated at 95±8 Bq/kg and 0.3, respectively. The radiation absorbed dose rates were estimated between 25±2 and 70±6 nGy/h and with an average of 44±4 nGy/h; however, the average annual effective dose of indoor gamma radiation, with the exception of the Radon, was calculated as 200±20 µSv/y for cumulative building materials.

The activity concentrations of the main naturally occurring radionuclides of <sup>40</sup>K, <sup>232</sup>Th, <sup>226</sup>Ra and <sup>235</sup>U were determined to be in the range of 426±20 to 740±28 Bq/kg (565±21 Bq/kg), 9±2 to 67±9 Bq/kg (25±5 Bq/kg), 9±1 to 31±3 Bq/kg (15±2 Bq/kg) and 0.03±0.01 to 0.7±0.1 Bq/kg (0.3±0.1 Bq/kg), respectively.

By taking into consideration of radiation as one of the primary factors caused cancer, the cumulative excess lifetime cancer risk of indoor radiation except radon, was calculated as 0.1±0.01% for the general population.

The figure 1 shows the average activity concentration of <sup>40</sup>K, <sup>226</sup>Ra and <sup>232</sup>Th (Bq/kg) in terms of each building material.



In the light of the results, the activity concentration index of all building material samples was calculated below the recommended value at 1 by Euroatom 2013.

In accordance with the Council Directive 2013/59/EURATOM, the samples are exempted for regular checking; in addition, the level of indoor external exposure to gamma radiation emitted by building materials was calculated to be below the recommended level as 1 mSv per year.

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